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Interception and Retention of Technetium by Vegetation and Soil[1]

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ABSTRACT

Field experiments were performed to quantify the initial interception and retention of technetium (Tc) by herbaceous vegetation as a consequence of direct deposition and uptake from soil. A simulated rain containing a solution of $^{95m}TcO_4^-$ was applied to plots of bare soil and plots with standing vegetation. Vegetation emerging in the bare-soil plots obtained Tc predominantly from root uptake. Vegetation standing during the initial application obtained Tc from both direct foliar interception and root uptake. For the plots with standing vegetation, the estimated initial interception fraction (r) ranged from 0.079 to 0.17. The mass interception factor (r/Y_v) ranged from 0.65 to 1.1 m²/kg. The retention of Tc by vegetation receiving direct foliar contamination varied with environmental half-times (T_w) ranging from 15.9 to 18.7 days. Adjusting for the effects of growth dilution increased the values of T_w (range 21.6–28.7 days). For vegetation emerging from the plots of initial bare soil, retention was equivalent to a T_w of 43 days. These data indicated that uptake of Tc from soil diminished with time and that Tc was removed from plant tissue. In soil, a downward movement was evident, because Tc in the top 2 cm decreased with time and concentrations at lower soil depths (6–8 and 14–15 cm) increased with time. The application of these data to predict steady-state concentrations of Tc in vegetation resulting from continuous deposition does not differ substantially from predictions based on the use of generic-default parameter values recommended in Regulatory Guide 1.109 of the U.S. Nuclear Regulatory Commission (1977).

Additional Index Words: **pertechnetate, radioecology, environmental assessment.**

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Technetium is a radioactive element of potential long-term importance in the environment. The long-lived isotope ^{99}Tc ($T_{1/2}$ = 212,000 years) exhibits a high rate of production during fission of nuclear fuels with yields of 6.1 and 6.4% from ^{235}U and ^{239}Pu, respectively. The pertechnetate anion (TcO_4^-) is highly soluble in water and poorly sorbed to soil, and thus is potentially mobile in biogeochemical cycles.

Laboratory studies have demonstrated exceptional bioaccumulation of Tc in vegetation (Cataldo et al., 1978; Gast et al., 1979; Routson and Cataldo, 1978; Mousny et al., 1979; Wildung et al., 1976, 1977, 1979). Vegetation grown in potted soils containing a solution of $^{99}TcO_4^-$ effectively removed up to 90% of the Tc from soil at concentrations below 0.1 μg/g. At soil concentrations above 0.1 μg/g the uptake was sufficient to produce toxic effects in vegetation (Wildung et al., 1975, 1977; Gast et al., 1979; Cataldo et al., 1977). Plant/soil concentration ratios (CR; ^{99}Tc/kg dry vegetation per ^{99}Tc/kg dry soil) from laboratory pot experiments are among the highest known for non-nutrient elements. Reported CR values range from 95 to 1,[illegible] for wheat seedlings (*Triticum aestivum* L. var. [illegible] (Gast et al., 1976); 67 to 380 for soybeans (*Glycine m*[illegible] cv. Williams) and wheat (*Triticum aestivum* [illegible] INTA66R) (Wildung et al., 1977); and 54 to 881 [illegible] cheatgrass (*Bromus tectorum*) and tumbleweed (*Salsola kali*) (Routson and Cataldo, 1978; Cataldo, 1979).

In the absence of field data, Till et al. (1979) demonstrated that the use of laboratory-derived plant/soil CR values for Tc in radiological assessment calculations would result in releases of ^{99}Tc from U-enrichment facilities potentially infringing on U.S. Environmental Protection Agency (EPA) standards for the U fuel cycle (USEPA, 1977). In their analyses they questioned the relevancy of laboratory data for such assessment calculations. The questions raised by Till et al. (1979) appear to be valid because recent field data on ^{99}Tc in soil and vegetation obtained near operating U-enrichment facilities show that field CR values are substantially less than those derived from laboratory pot experiments (Hoffman et al., 1980). These field CR values ranged from 1.4 to 44, with a geometric mean of 9.5.

Prior to the present study there were no field data on the interception and retention of Tc deposited on and incorporated within vegetation. Typically, this information is not obtained from laboratory pot experiments. Vegetation grown in the lab on potted soils is usually protected from factors normally prevalent in the field, such as wind, rain, and grazing, which could reduce the Tc inventory in field vegetation (Tukey, 1970; Beauford et al., 1977; Batzli, 1978). Furthermore, no attempt has been made in the lab experiments to simulate the effects of direct deposition whereby the soil and vegetation are simultaneously exposed to Tc.

A recent review by Miller (1980) summarizes the results of experiments on the interception of a variety of radioactive and other aerosols by vegetation. A strong correlation is evident between the fraction of deposited aerosols initially intercepted by standing forage vegetation (r) and standing crop biomass (Y_v; kg/m² dry wt). The range of observed interception fractions for forage vegetation is from 0.02 to 0.82, while the range of values representing the observed ratio of the interception fraction to standing crop biomass (r/Y_v) is from 1.0 to 4.0 m²/kg. The observed ratio r/Y_v is often referred to as a "mass interception factor" since the product of this ratio and the total area deposition is the concentration per mass vegetation. The observed ratio r/Y_v is determined experimentally by dividing the measured concentration per mass vegetation by the total area deposition.

Miller and Hoffman (1979) reviewed a number of field studies on the retention by herbaceous vegetation

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of a variety of radioisotopes deposited as aerosols. Correcting for radioactive decay, the environmental half-times (T_w) associated with the retention of these materials range from 8.7 to 27 days during the growing season, with a median value of 13.4 days. A winter value of 71 days for a solution of $^{85}SrCl_2$ sprayed on grassland is also reported in this review. Particulate deposition on desert shrubs exhibits values of T_w ranging from 13 to 28 days. No indication of the possible effect of dilution of activity by increasing biomass during vegetation growth is given.

It is generally assumed that the values of T_w are relevant only to the retention of aerosols initially deposited on the plant surfaces. The degree to which these values include the retention by vegetation of material taken up from soil and incorporated within plant tissue is not specified. Although the environmental half-time is often presumed to represent removal due to weathering, correlations between T_w and specific meteorological variables such as precipitation and wind generally have not been confirmed. Arkhipov and Fevraleva (1980) showed that the foliar content of ^{89}Sr, applied as a solution to a variety of crops, was inversely correlated with post-contamination rainfall.

Most studies on the retention of Tc in soil have been confined to laboratory experiments of the distribution coefficient (K_d; the ratio of the amount sorbed per gram soil to the amount in solute per milliliter equilibrating solution). Under aerobic conditions, Wildung et al. (1979) report K_d values for pertechnetate (equilibrated for 24 hours) that range from 0.007 to 2.8, indicating the potential for high mobility in soil. However, 98% sorption has been observed for equilibrating periods of 2-5 weeks and for soil conditions tending to be anaerobic (Landa et al., 1977). An estimate of the removal rate of TcO_4^- from the root zone of soils has been calculated by Baes (1979), in which the loss rate is considered to be a function of the velocity of vertical water percolation in soil, depth of the soil root zone, soil bulk density, soil water content, and values of K_d. Using median values for all parameters, Baes calculated a median-rate constant for the leaching of TcO_4^- from the 0- to 15-cm root zone of soil of 2.6/year, equivalent to an environmental half-time of about 100 days. Because of the large variability in reported values of the velocity of vertical water percolation and in K_d, Baes emphasizes the uncertainty associated with his estimate and recommends field measurements to confirm his results.

Without available information specific to Tc interception and retention, the present version of the environmental assessment model used by the U.S. Nuclear Regulatory Commission (NRC) assumes that initial interception (r) is 20–25% (USNRC, 1977). Equivalent values of the mass interception factor (r/Y_v) derived from the NRC model are 1.4 m²/kg for irrigation spray and 1.1 m²/kg for particulates. These values are derived from default values of r and Y_v used by the model in lieu of element-specific or site-specific data. Retention of materials deposited directly on the surfaces of vegetation is assumed to be equivalent to an environmental half-time (T_w) of 14 days. In soil, the NRC model adopts the pessimistic assumption that all deposited material will eventually reside in the root zone of soil (0-to 15-cm depth), with the only source of removal being radioactive decay. Retention in soil is, therefore, assumed to be complete.

In this study, field experiments were designed to record the behavior of Tc in soil and vegetation subsequent to a single spike application. The objectives of this study were to obtain data relevant to field conditions that could be compared with interception and retention values assumed in environmental assessment models. Estimates of vegetation retention of Tc were made from observations of the initial contamination of the soil as well as direct deposition onto plant surfaces in order to include the combined effects of root uptake and removal of Tc from plant tissue. The retention of Tc in soil was estimated through observations of Tc concentrations at various depths over time subsequent to initial deposition.

MATERIALS AND METHODS

Field experiments were conducted in a 2,500-m² old field within the 0800 area of the U.S. Department of Energy Oak Ridge Reservation, Oak Ridge, Tenn. The soil in this field is a Captina silt loam (typic Fragiudult) composed of 13.7% sand, 75.2% silt, and 11.1% clay by weight. Soil organic matter is about 6–8%. The soil cation exchange capacity is 14.4 meq/100 g, and the pH is 5.0.

Prior to the establishment of experimental plots, the old field was mechanically mowed to enhance dominance of herbaceous vegetation. Fescue grass (*Festuca arundinacea*) was the most abundant cover vegetation present after mowing, with sericea lespedeza (*Lespedeza cuneata*) occurring in increasing abundance during the latter phases of each experiment. Technetium was applied once to each plot as a solution of carrier-free $^{95m}TcO_4^-$ sprayed (600- to 1,500-μm diameter droplets) by hand through a perforated lid attached to a 1-liter polyethylene bottle. Care was exercised to maintain uniform application over each experimental plot. The radionuclide and radiochemical purity of the ^{95m}Tc was 99%.

To provide an estimate of the retention of Tc in vegetation following root uptake from soil, 15 1-m² plots were randomly selected in which all vegetation was removed to expose bare soil prior to the application of ^{95m}Tc (hereafter these plots will be referred to as the bare-soil plots). Each plot received approximately 200 μCi of ^{95m}Tc. Nine days after application of ^{95m}Tc, a 2-cm diameter soil core was

Table 1—Sampling schedule and accumulated rain during each field experiment.

Bare-soil plots (26 Sept. 1978–27 Apr. 1979)		Foliar Plot A (25 Apr. 1979–30 July 1979)		Foliar Plots B, C, D (17 July 1979–19 Oct. 1979)	
Sample day†	Rain‡	Sample day†	Rain‡	Sample day†	Rain‡
9	1.27§	0	1.27¶	0	0.00¶
14	0.76§	1	1.30	1	0.00
20	0.00	2	0.00	2	0.00
24	0.00	5	0.23	6	12.67
41	1.52§	6	0.00	13	8.56
57	9.63	7	0.00	20	0.00
84	14.99	16	4.50	31	4.09
148	27.99	26	3.35	41	5.11
171	13.74	34	9.04	50	5.21
213	17.58	40	13.16	66	5.03
Total	87.48	47	0.41	80	4.57
		65	2.08	94	2.36
		83	12.19	Total	47.60
		96	21.23		
		Total	68.76		

† Time of sample in days since ^{95m}Tc applied to experimental field plots.
‡ Rainfall accumulated since time of previous sample.
§ Rainfall simulated by hand watering. Thirteen liters of water per plot applied on 2 Oct. 1978; 7.6 liters of water per plot applied on 9 Oct. 1978, 25 Oct. 1978, and 30 Oct. 1978. One centimeter of rain is equivalent to liters/m² plot. Plastic tents removed from plots on 3 Nov. 1978.
¶ Rainfall accumulated during the day in which ^{95m}Tc was applied to experimental field plots.

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taken to a depth of 15 cm from each plot. The cores were segmented into 0- to 1-, 1- to 2-, 6- to 8-, and 14- to 15-cm depths. Soil samples were taken during 10 sampling periods at intervals of decreasing frequency during the 213-day duration of the experiment (Table 1).

Secondary growth vegetation emerging from the bare soil was not sampled until after the 20th day post-application, at which time sufficient biomass, composed primarily of fescue grass, had accumulated within the plots to permit collection. Vegetation was then removed from each plot by clipping small quantities (0.1–0.3 g, dry wt) of living green tissue within the plot at several random locations. Subsequent vegetation samples were taken at the same time as the soil cores. The plots were covered by clear plastic tents for the first 5 weeks of the experiment to prevent contamination of emerging vegetation by rain splash. During this time the plots were watered artificially by a light spray of distilled water to an amount prescribed by ambient rainfall (Table 1). The duration of the experiment was from 26 Sept. 1978 to 27 Apr. 1979.

To estimate the effective retention of Tc in vegetation after direct deposition onto the leaves and subsequent root uptake of Tc deposited in the soil, as well as initial interception of deposited Tc by vegetation, four 1-m² plots were selected in which the standing vegetation was not disturbed after initial mowing (hereafter these plots will be referred to as foliar plots). Each plot received approximately 10 μCi of ^{95m}Tc. The experiment was conducted in two series with slight modifications in sampling technique. In the first series a single plot (Foliar Plot A) was studied from 25 Apr. 1979 to 30 July 1979. Three vegetation samples composed mainly of fescue were obtained at random from the plot during each sampling period. Standing biomass was estimated by clipping vegetation to a height of 2 cm above the soil from three adjacent 400-cm² areas outside the plot, and oven-drying at 70°C prior to weighing.

Six sampling periods occurred during the 1st week, with eight sampling periods occurring at decreasingly frequent intervals throughout the remainder of the 96-day experiment (Table 1). After the first 6 weeks of the experiment, three soil cores were obtained at the same intervals as for vegetation. The first samples of soil were taken from the 0- to 2-cm soil depth only. The final four samples of soil included the same depth intervals as were taken from the 15 bare-soil plots. Two replicate samples of *Lespedeza* were taken in addition to samples of fescue grass, as the result of the appearance of the former 1 month after initial application of Tc.

In the second series, three replicate plots (Foliar Plots B, C, and D) were studied from 17 July 1979 to 19 Oct. 1979. Three samples of vegetation and soil were obtained during each sampling peirod. As with the initial bare-soil plots, soil cores were taken at the 0- to 1-, 1- to 2-, 6- to 8-, and 14- to 15-cm depths. Four sampling periods occurred during the 1st week of the experiment, followed by eight sampling periods occurring at decreasingly frequent intervals (Table 1). During each sampling period, standing biomass was estimated in a similar manner as for Foliar Plot A. At the termination of this experiment, plant-species differences in Tc concentration were determined by sampling five distinct species at five locations within each of the three plots.

For each foliar plot, the mass interception factor (r/Y_v) was obtained directly by dividing the Tc concentration per mass vegetation (taken 1 hour after application of $^{95m}TcO_4^-$) by the total known amount of Tc applied to the plot. The interception fraction (r) was then obtained by multiplying the mass interception factor (r/Y_v) by the estimated standing biomass (Y_v) present at the time of Tc application:

$$r = (r/Y_v)(Y_v). \qquad [1]$$

Sample preparation and counting procedures were the same for the foliar plots as for the bare-soil plots. All fresh samples were placed in preweighed counting tubes and counted using gamma spectroscopy in a deep-well multichannel Na-I crystal linked to a pulse height analyzer (PHA). Soil and plant tissue were subsequently dried in a forced-draft oven at 70°C for 24 hours, and weighed. All measurements of radioactivity in vegetation and soil were corrected for radioactive decay of ^{95m}Tc (61-day half-life). All data were subject to analysis using a statistical package, SAS (Helwig and Council, 1979), from which linear regressions of logtransformed observations were used to estimate retention of Tc in vegetation and soil with time.

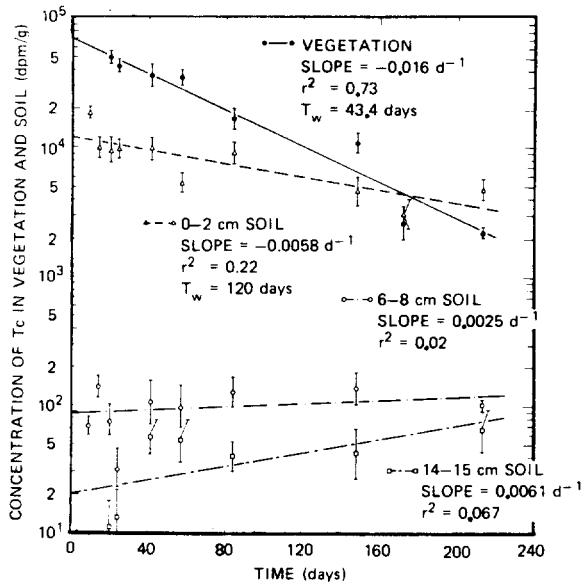

Fig. 1—Geometric means and standard errors (n = 15) of decay-corrected ^{95m}Tc concentrations in vegetation and soil. Technetium was applied as a simulated light rain directly to 15 1-m² plots of initial bare soil.

RESULTS

In the 15 bare-soil plots, the concentrations of Tc in emerging vegetation decreased with time (Fig. 1). Retention of Tc in vegetation was approximated by an effective first-order rate constant of 0.016 days^{-1} (±0.00091 SE). This effective rate constant was equivalent to an environmental half-time of 43 days and most probably included the combined processes of continuous uptake of Tc from soil, dilution of Tc in vegetation from the addition of plant biomass during growth, and physical removal of Tc via weathering from vegetation. Growth dilution was presumed to be minimal because winter conditions inhibited growth from the 3rd to the beginning of the 7th month. No direct measurement of standing-crop biomass was made, due to the sparse re-

Table 2—Effective first-order rate constants, λ_w (corrected for ratioactive decay), and associated environmental half-times T_w, for secondary-growth vegetation in which $^{95m}TcO_4^-$ was initially applied as simulated rainfall to bare soil.

Plot	Rate constant λ_w (day^{-1})	Environmental half-time T_w (day)	r^2†
1	0.0135	51.3	0.885**
2	0.0140	49.5	0.929**
3	0.0175	39.6	0.818**
4	0.0171	40.5	0.836**
5	0.0129	53.7	0.649*
6	0.0140	49.5	0.903**
7	0.0181	38.3	0.888**
8	0.0128	54.2	0.477
9	0.0198	35.0	0.796**
10	0.0185	37.5	0.621*
11	0.0197	35.2	0.906**
12	0.0115	60.3	0.559
13	0.0188	36.9	0.955**
14	0.0115	60.3	0.885**
15	0.0176	39.4	0.755**

* $P < 0.05$.
** $P < 0.01$.
† r^2 = coefficient of determination.

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covery of vegetation in the plots. Nevertheless, results for each of the 15 plots illustrate a consistency in the effective loss of Tc from vegetation (Table 2).

In soil, a downward movement of Tc was evident. A significant ($P < 0.01$) loss of Tc was observed from the top 0-2 cm of soil with increases in Tc concentrations observed at the 6- to 8- and 14- to 15-cm soil depth (Fig. 1). The effective first-order rate constant calculated for the loss of Tc in the top centimeter of soil was 0.0065 day^{-1} (± 0.009 SE), equivalent to an environmental half-time of 106 days. The rate constant calculated for the 0- to 2-cm soil depth was 0.0058 day^{-1} (± 0.0009 SE), equivalent to an environmental half-time of 120 days.

The amount of Tc sampled at the 0- to 2-, 6- to 8-, and 14- to 15-cm soil depths was determined by multiplying the activity concentration (dpm/g) by the bulk density for the intact soil at a specific sampling depth (g/cm^3), and by the volume of the sample taken from the core (3.14 cm^2 × length [cm] of the sample segment). The soil bulk densities for the three sampling depths were 1.21, 1.57, and 1.57 g/cm^3, respectively.

Linear interpolation between the amount of Tc sampled at the 0- to 2-, 6- to 8-, and 14- to 15-cm soil depths produced an estimate of the total Tc in the 0- to 15-cm soil core. Regression of the total core Tc with time resulted in a rate constant approximating the effective rate of removal of Tc from the 0- to 15-cm root zone of 0.00365 day^{-1} (± 0.001 SE), equivalent to an environmental half-time of 190 days. Negative correlations between soil Tc and time were small but significant ($r = -0.31$, $P < 0.01$), due to large between-plot variability. Uptake by vegetation had little effect on the removal of Tc from the soil because the total Tc in vegetation was <0.3% of the total Tc estimated in the 0- to 15-cm root zone of soil.

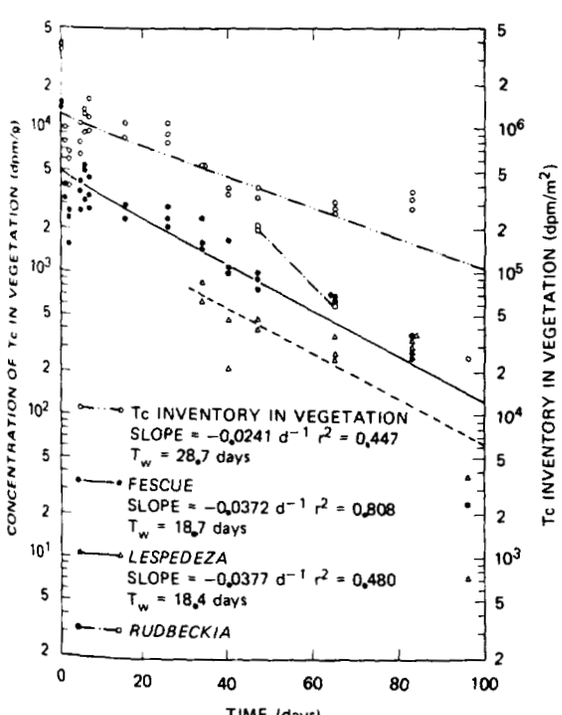

Fig. 2—Decay-corrected ^{95m}Tc concentration and inventory in vegetation of Foliar Plot A. Each point represents the result for an individual sample of vegetation at a given time period.

Table 3—Values of estimated interception fractions (r) and mass interception fractions (r/Y_v) obtained from Foliar Plots A–D.

Plot	Tc/m² (initial application)	Tc/g vegetation (at $T = 1$ hour)	r/Y_v	Biomass	r
	dpm	dpm	m²/kg	kg/m²	
A	2.22×10^7	1.45×10^4	0.652	0.254	0.166
B	2.22×10^7	2.24×10^4	1.01	0.078	0.079
C	2.22×10^7	2.46×10^4	1.10	0.135	0.150
D	2.22×10^7	2.40×10^4	1.08	0.135	0.146
Combined (B,C,D)	2.22×10^7	2.37×10^4 $\pm 0.11 \times 10^4$	1.07 ± 0.05	0.116 ± 0.003	0.125 ± 0.04

The initial interception fraction (r) and the mass interception factor (r/Y_v) for each foliar plot are presented in Table 3. The estimated initial interception (r) ranged from 0.079 to 0.166; the mass interception factor (r/Y_v) ranged from 0.652 to 1.10 m^2/kg. The lower value of r/Y_v for Foliar Plot A was measured after a light rain that occurred immediately after application of TcO_4^-.

In the foliar plots, both the concentration and the total amount of Tc in vegetation decreased with time (Fig. 2 and 3). As in the previous experiments, these results indicated a physical removal of Tc from vegetation. Some influence of growth dilution on estimates of the environmental half-time T_w was evident. Values of T_w obtained from regressions of the concentration of Tc per mass vegetation included the effects of growth dilution. These values were consistently shorter than values of T_w that were obtained from regressions of the concentration of Tc in vegetation per unit area of ground. Values of T_w based on regressions of the inventory of Tc in vegetation effectively exclude the effect of growth dilution (Table 4). Values of the environmental half-time (T_w) that include growth dilution ranged from 15.9 to 18.7 days; values of T_w that exclude growth dilution ranged from 21.6 to 28.7 days.

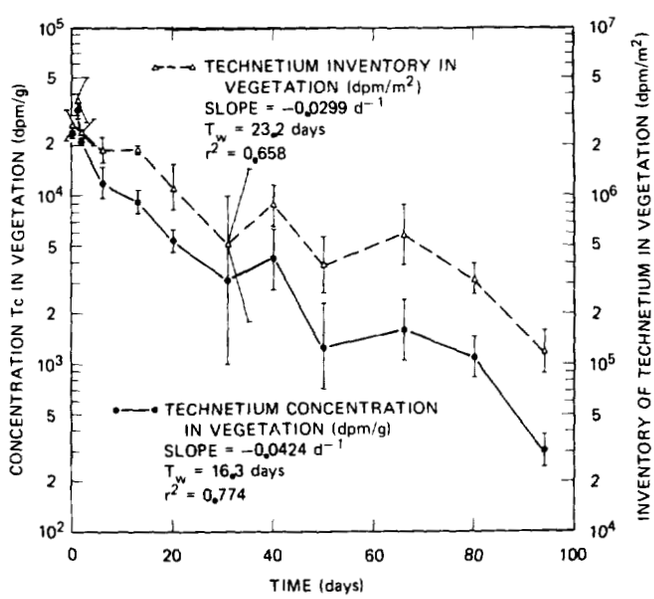

Fig. 3—Geometric means and standard errors ($n = 3$) of decay-corrected ^{95m}Tc concentration and inventory in vegetation for Foliar Plots B, C, and D.

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Table 4—Values of effective first-order weathering rate constants (λ_w) and corresponding environmental half-times (T_w) describing the retention of Tc by foliar plot vegetation.

Plot	λ_w concn.†	T_w concn.†	r^2§	λ_w inv.‡	T_w inv.‡	r^2
	day^{-1}	days		day^{-1}	days	
A	0.0372	18.7	0.808**	0.0241	28.7	0.447*
B	0.0410	16.9	0.904**	0.0274	25.3	0.857**
C	0.0436	15.9	0.811**	0.0322	21.6	0.735**
D	0.0424	16.3	0.687**	0.0300	23.1	0.532**
Combined (B,C,D)	0.0424	16.3	0.775**	0.0300	23.1	0.657**

* $P < 0.05$.
** $P < 0.01$.
† Concn. refers to values obtained from regressions against time of decay-corrected Tc concentration in vegetation (dpm/g).
‡ Inv. refers to values obtained from regressions against time of decay-corrected Tc inventory in vegetation (dpm/m²).
§ r^2 = coefficient of determination.

Species of vegetation in the foliar plots that were not evident at the time of initial deposition of Tc also exhibited a reduction of Tc concentration with time. *Lespedeza* sampled in Foliar Plot A, upon its appearance 1 month after initial application (Fig. 2), lost Tc at a rate comparable to that estimated for fescue. The environmental half-time (T_w) for the decay-corrected concentration of Tc in *Lespedeza* was 18.4 days. Two leaf samples from a black-eyed Susan (*Rudbeckia hirta*) within Foliar Plot A also indicated a loss of Tc from the plant (Fig. 2).

Sampling of Tc concentrations in herbaceous vegetation in Foliar Plots B, C, and D at the end of the experiment revealed considerable species variation (Table 5). This variability may reflect true differences among species, or differences related to the time after the initial application of Tc when different species appeared in each plot. It is possible that new growth for all species was sufficiently abundant so that most samples taken at the end of the experiment did not contain surface contamination of Tc from direct deposition at time zero. The large increase in the variability of vegetation concentrations sampled among all plots, about 1 month after initial Tc application (Fig. 3), coincided with the appearance of new growth and successional growth vegetation.

With the exception of Foliar Plot A, no correlation was found between the retention of Tc by vegetation and the amount of precipitation accumulated between each sampling period. Accumulated rainfall for each experiment is given in Table 1. For Foliar Plot A, a small ($r = -0.57$) but significant ($P < 0.05$) negative correlation was determined between retention of Tc and accumulated rainfall. Removal of Tc from vegetation by precipitation is suspected because simulated rainfall has been demonstrated to be effective in removing Tc from vegetation under laboratory conditions in which the roots of the plants accumulated $^{99}TcO_4^-$ from contaminated Hoagland's solution (Myttenaere et al., 1980).

Technetium in soil samples taken from Foliar Plot A were not significantly correlated with time ($P > 0.05$), although the concentration of Tc in the top 2 cm tended to exhibit lower values towards the end of the experiment (Fig. 4). Soil samples from Foliar Plots B, C, and D were taken over a substantially longer period of time (94 days) than samples from Foliar Plot A (56 days).

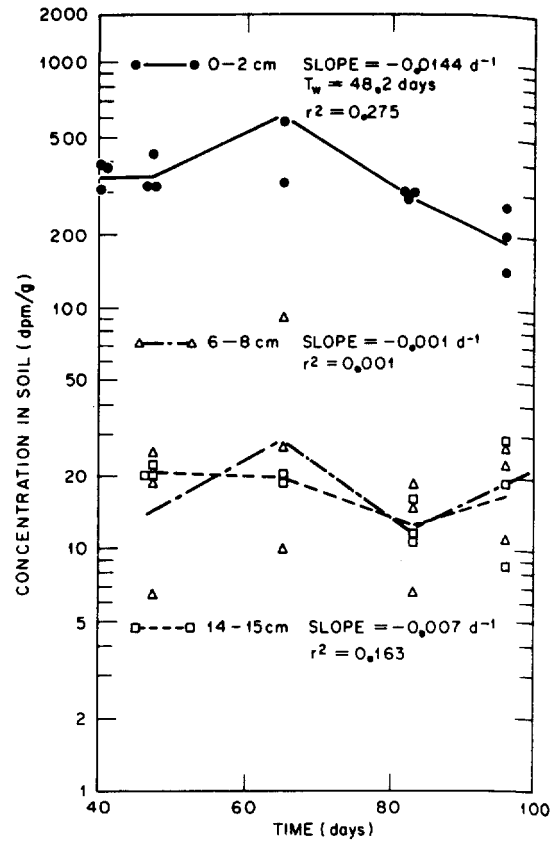

Fig. 4—Decay-corrected concentration of ^{95m}Tc in various soil depths within Foliar Plot A. Each point represents the result for an individual soil sample at a given time period.

Therefore, samples pooled among Foliar Plots B, C, and D indicated a significant decrease of Tc ($P < 0.05$) with time in the top 2 cm of soil and a significant ($P < 0.01$) increase in Tc concentration at depths of 6–8 and 14–15 cm (Fig. 5). For Foliar Plots B, C, and D, the effective first-order rate constant calculated for the loss of Tc in the top centimeter of soil was 0.00899 day^{-1} (± 0.0023 SE), equivalent to an environmental half-time of 78 days. The rate constant calculated for the 0- to 2-cm soil depth was 0.0045 day^{-1} (± 0.0017 SE), equivalent to an environmental half-time of 156 days. Significant trends ($P < 0.05$) for the concentration of Tc within the entire 0- to 15-cm soil profile of these foliar plots could not be detected, perhaps due to insufficient length of the experiment.

DISCUSSION

The results from the application of $^{95m}TcO_4^-$ to the bare-soil plots (Fig. 1) and the foliar plots (Fig. 2 and 3) appear to indicate that Tc is physically removed from vegetation, regardless of whether the Tc content in vegetation is from direct deposition onto plant surfaces or root uptake from soil and translocation into aboveground portions of the plant. In addition, decreasing availability of Tc in soil for plant uptake, due to its transformation to less soluble forms, in combination with growth dilution could explain some of the decrease in plant Tc concentrations over time. The prevalence of an apparent constant rate in which Tc was lost from vegetation also implies either that uptake of Tc from

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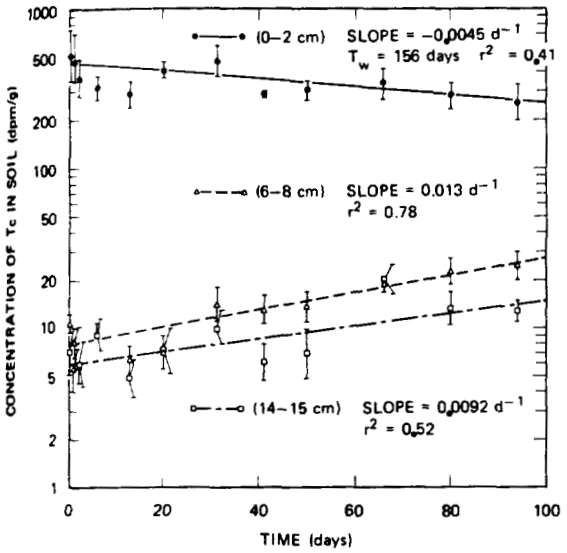

Fig. 5—Geometric means and standard errors (n = 9) of decay-corrected concentrations of ^{95m}Tc at various soil depths for Foliar Plots B, C, and D.

soil decreases with time or that the removal of Tc from vegetation increases with time. We suspect the former, as reduction in soil of the soluble $Tc(VII)O_4^-$ to less soluble, lower oxidation states of Tc(VI, V, and/or IV) would account for diminished root uptake. Chemical reduction is suspected because in the laboratory only 33% of the Tc in soil could be leached using distilled water. Of that remaining, only 2% was extracted with 0.1N NaCl. However, a cold 5% H_2O_2 solution, which would oxidize Tc(IV) to Tc(VII), removed 46% more of the soil Tc.

Leaching by precipitation is the mechanism assumed to remove Tc from vegetation, although the release from vegetation of submicrometer-sized particles during periods of high transpiration and rapid growth, as well as the release of larger particles induced by wind action, is also possible (Beauford et al., 1977). Leaching of inorganic substances from vegetation is a recognized phenomenon, but loss rates may differ with the age of the plant as well as the intensity and volume of precipitation (Tukey, 1970). According to Tukey, mature vegetation is more susceptible to leaching than new growth, and rain falling as a light continuous drizzle is more efficient as a leaching agent than is a large quantity of rain falling during a short period. No attempt was made in this study to specify the maturity of vegetation sampled or the specific type, intensity, and volume of precipitation received by vegetation. The loss rate of Tc from vegetation could not be explained by a simple function of rainfall accumulated between sampling periods.

Table 5—Concentrations of Tc in various species of herbaceous vegetation.†‡

Vegetation	Geometric mean and (geometric SD)			
	Foliar Plot B	Foliar Plot C	Foliar Plot D	All plots combined
	dpm/g			
Lactuca	243 (1.92)	54.5 (2.79)	125 (1.69)	125[a] (2.50)
Lespedeza	678 (1.66)	426 (4.60)	218 (1.96)	398[b] (2.85)
Panicum	960 (1.46)	278 (1.45)	822 (4.19)	605[b] (2.72)
Sorghum	35.3 (3.28)	33.3 (1.51)	17.6 (1.73)	28.5[d] (2.17)
Sorbus	94.4 (2.62)	– –	124 (2.76)	108[a] (2.57)

† Values with the same alphameric superscript are not statistically different ($P > 0.05$).
‡ Vegetation collected on 19 Oct. 1979, at five locations within each plot.

In general, a stronger correlation occurred between time and the concentration of Tc in vegetation (dpm/g), than between time and the inventory of Tc in vegetation (dpm/m²). This stronger correlation is because the process of growth dilution is included in regressions of Tc concentration in vegetation (dpm/g) and time. Values of effective half-time, T_w (15.9–18.7 days) that include the process of growth dilution (Table 3) are comparable to the median value of T_w (13.4 days) reported in the literature for the retention of radioactive aerosols by herbaceous vegetation during the growing season (Miller and Hoffman, 1979). Values of T_w (21.6–43.4 days) based on the Tc inventory in vegetation (Table 3) and dormant winter-growth vegetation (Fig. 1) are more comparable to the maximum values of T_w reported in the literature for radioactive aerosols.

The conventional use of T_w for vegetation in assessment models assumes that removal of radioactive substances occurs primarily from plant surfaces. The results obtained in this study only partially support this assumption. The effective environmental half-times T_w obtained from these field experiments include the combined effects of physical removal of Tc from vegetation, uptake of Tc from soil, and dilution of Tc by tissue growth.

Vegetation/soil concentration ratios (CR), determined by dividing the Tc concentration in vegetation by the Tc concentration in the top 2 cm of soil at approximately 50 days after the initial time of Tc application, were calculated for each field experiment in order to make comparisons between plots having different modes of contamination. It should be noted, however, that CR values will vary if the depth of soil chosen for the calculation changes. A median CR value derived from the 15 plots of initial bare soil (Fig. 1) was about 2.5; for Foliar Plot A (Fig. 2 and 4), approximately 4.2; and for the combined samples of vegetation and soil from Foliar Plots B, C, and D (Fig. 3 and 5), approximately 2.6. This similarity in CR values is of interest be-

Table 6—The predicted concentration of Tc in vegetation resulting from a continuous deposition rate of Tc using values of r/Y_v and T_w obtained from the Foliar Plots A, B, C, and D and default values recommended by the U.S. NRC.

Foliar Plot A	Foliar Plot B	Foliar Plot C	Foliar Plot D	NRC
Assumed deposition rate, $\dot{d}$ (dpm/m² · day)				
1.0	1.0	1.0	1.0	1.0
Mass interception factor, r/Y_v (m²/kg)				
0.652	1.01	1.10	1.08	1.1†
Environmental half-time, T_w (days)				
18.7	16.9	15.9	16.3	14†
Predicted concentrations of Tc in vegetation at equilibrium, C_v (dpm/kg)§				
17.6	25.6	25.2	25.4	22.2‡

† NRC value assumed for aerosol deposition.
‡ Excludes the process of uptake from soil.
§ Equilibrium concentration of Tc in vegetation (C_v) is calculated as $C_v = \dot{d} \cdot r/Y_v \cdot T_w/\ln 2$.

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cause vegetation that grew into the initial bare-soil plots obtained Tc entirely through root uptake, while vegetation in the foliar plots received Tc through both direct deposition onto above-ground plant surfaces and root uptake from soil. Root uptake of Tc from soil is considered to contribute significantly to the total Tc content in vegetation for each field experiment, especially since much of the vegetation sampled in the later phases of the foliar plot experiment was not present at the time of initial Tc application.

Estimates (Table 2) of the mass interception factor (r/Y_v) and the initial interception fraction (r) are near the lower end of the range of values reported in the literature for radioactive aerosols and fine water droplets (Miller, 1980). Factors influencing these results might be related to the intensity of the spray in which $^{95m}TcO_4^-$ was initially applied to the foliar plots, the growth form of the vegetation and prevailing meteorological conditions at the time of application, and the amount of standing biomass in each foliar plot. Nevertheless, the results obtained in this study for interception and retention of Tc by vegetation are comparable to the generic-default values recommended in the environmental radiological assessment models of the NRC Regulatory Guide 1.109 (USNRC, 1977). The predicted concentration of Tc in vegetation at equilibrium (Table 6) is only slightly different, depending on whether values of r/Y_v and T_w are taken from this study or from the generic default recommendations given in the NRC Regulatory Guide.

Unlike the assumption in Regulatory Guide 1.109 (USNRC, 1977) that no loss of Tc other than radioactive decay will occur in the 0- to 15-cm root zone of soil over a time period of 15 years, the results (Fig. 1 and 5) suggest a downward migration of Tc in soil. The median prediction for a leaching-rate constant for TcO_4^- of 2.6 years^{-1} estimated by Baes (1979) is comparable to values (2.1 and 1.6 years^{-1}) obtained for the top 2 cm of soil. Baes' estimate, however, is specific for the 0- to 15-cm root zone. The estimated effective first-order rate constant for the loss of Tc from the 0- to 15-cm root zone of soil for the 15 bare-soil plots was approximately 1.3 year^{-1} ($\pm$0.36 SE). This is a factor of two lower than Baes' predicted median value. The NRC assumption of no loss of Tc from the soil root zone is conservative and would result in predicted soil concentrations of ^{99}Tc after 15 years of continuous deposition approximately 20 times greater than would occur if the movement of Tc out of the 0- to 15-cm root zone could be characterized by a rate constant of 1.3 years^{-1}.

The relevance of the data on interception and retention of Tc obtained from these experiments to situations involving prolonged releases of ^{99}Tc remains to be tested through field validation studies. Different soil types and vegetation species, and longer periods of exposure to ^{99}Tc, could potentially produce results that deviate from the trends reported in this study. Comparable results, however, have been produced between measured vegetation/soil CR values[5] for ^{99}Tc monitored near operating gaseous-diffusion plants, and estimated steady-state CR values calculated using data reported herein on interception and retention of ^{95m}Tc.

[5]F. O. Hoffman, C. T. Garten, Jr., D. M. Lucas, and J. W. Huckabee, 1981. Environmental behavior of technetium in soil and vegetation—implications for radiological assessment. Environ. Sci. Technol. (submitted). Note that the CR values referred to in this reference are based on soil concentrations in the 0- to 15-cm root zone and are therefore different than values reported in this manuscript.

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LITERATURE CITED

1. Arkhipov, N. P., and L. T. Fevraleva. 1980. Role of rainfall in the elimination of radionuclides from plants after their foliar contamination. Sov. J. Ecol. 10:530–534. Translated into English from Ekologiya 10:41–46.
2. Baes, C. F. III. 1979. The soil loss constant λ_{sl} due to leaching from soils. p. 85–92. F. O. Hoffman and C. F. Baes, III (ed.) A statistical analysis of selected parameters for predicting food chain transport and internal dose of radionuclides. Oak Ridge Natl. Lab., Pub. ORNL/NUREG/TM282. Natl. Tech. Inf. Serv., Springfield, Va.
3. Batzli, G. O. 1978. The role of herbivores in mineral cycling. p. 95–112. *In* D. C. Adriano and I. Brisbin (ed.) Environmental chemistry and cycling processes. CONF-760429. Natl. Tech. Info. Serv., Springfield, Va.
4. Beauford, W., J. Barber, and A. R. Barringer. 1977. Releases of particles containing metals from vegetation to the atmosphere. Science 195:571–573.
5. Cataldo, D. A. 1979. Behavior of technetium and iodine in a hanford sand and associated subsoil: influence of soil aging on uptake by cheatgrass and tumbleweed. U.S. Dep. of Energy Rep. PNL2740. Natl. Tech. Info. Serv., Springfield, Va.
6. Cataldo, D. A., R. E. Wildung, and T. R. Garland. 1978. Technetium accumulation, fate, and behavior in plants. p. 538–549. *In* D. C. Adriano and I. L. Brisbin, Jr. (ed.) Environmental chemistry and cycling processes. Natl. Tech. Info. Serv., Springfield, Va.
7. Gast, R. G., E. R. Landa, and L. J. Thorvig. 1976. The behavior of technetium-99 in soils and plants: final report for the period April 1, 1974, to March 31, 1977. C00-2447-5. Natl. Tech. Info. Serv., Springfield, Va.
8. Gast, R. G., E. R. Landa, L. J. Thorvig, D. F. Grigal, and J. C. Balogh. 1979. The behavior of technetium-99 in soils and plants: final report for the period April 1, 1974, to December 31, 1978. C00-2447-6. Natl. Tech. Info. Serv., Springfield, Va.
9. Helwig, J. T., and K. A. Council. 1979. SAS user's guide. Statistical Analysis System Institute, Inc., Raleigh, N.C.
10. Hoffman, F. O., J. W. Huckabee, D. M. Lucas, C. T. Garten, T. G. Scott, R. L. Walker, P. S. Gouge, and C. V. Holmes. 1980. Sampling of technetium-99 in vegetation and soils in the vicinity of operating gaseous diffusion facilities. Oak Ridge Natl. Lab. Rep. ORNL/TM-7386. Natl. Tech. Info. Serv., Springfield, Va.
11. Landa, E. R., L. H. Thorvig, and R. G. Gast. 1977. Effect of selective dissolution, electrolytes, aeration, and sterilization on technetium-99 sorption by soils. J. Environ. Qual. 6:181–187.
12. Miller, C. W. 1980. An analysis of measured values for the fraction of a radioactive aerosol intercepted by vegetation. Health Phys. 38:705–712.
13. Miller, C. W., and F. O. Hoffman. 1979. The environmental loss constant for radionuclides deposited on the surfaces of vegetation, λ_w. p. 43–50. *In* F. O. Hoffman and C. F. Baes, III (ed.) A statistical analysis of selected parameters for predicting food chain transport and internal dose of radionuclides. Oak Ridge Natl. Lab. Rep. ORNL/NUREG/TM-282. Natl. Tech. Info. Serv., Springfield, Va.
14. Mousny, J. M., P. Roucoux, and C. Myttenaere. 1979. Absorption and translocation of technetium in pea plant. Environ. Exp. Bot. 19:263–268.
15. Myttenaere, C., C. Daoust, and P. Roucoux. 1980. Leaching of technetium from foliage by simulated rain. Environ. Exp. Bot. 20:415–419.
16. Routson, R. C., and D. A. Cataldo. 1978. Accumulation of ^{99}Tc by tumbleweed and cheatgrass grown on arid soils. Health Phys. 34:685–690.
17. Till, J. E., F. O. Hoffman, and D. E. Dunning, Jr. 1979. A new

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look at ^{99}Tc releases to the atmosphere. Health Phys. 36:21-30.
18. Tukey, H. B., Jr. 1970. The leaching of substances from plants. Annu. Rev. Plant Physiol. 21:305-324.
19. U.S. Environmental Protection Agency. 1977. Radiation protection programs. Environmental radiation protection standards for nuclear power operations. Fed. Regist. 42(9):2858-2861.
20. U.S. Nuclear Regulatory Commission. 1977. Calculation of annual doses to man from routine releases of reactor effluents for the purpose of evaluating compliance with 10 CFR Part 50, Appendix I. Regulatory Guide 1.109, Revision 1, 1 Oct. 1977. NRC, Washington, D.C.
21. Wildung, R. E., D. A. Cataldo, and T. R. Garland. 1975. Accumulation of Tc from Soil by Plants. USERDA Rep. BNWL-2000. Part 2. Natl. Tech. Info. Serv., Springfield, Va. p. 40-42.
22. Wildung, R. E., D. A. Cataldo, and T. R. Garland. 1976. Accumulation of Tc from Soil by Plants. I. Uptake of Technetium from Soil by Soybeans and Wheat. USERDA Rep. BNWL-2000. Pa t 2. Natl. Tech. Info. Serv., Springfield, Va. p. 37-39.
23. Wildung, R. E., T. R. Garland, and D. A. Cataldo. 1977. Accumulation of technetium from soil by plants. Health Phys. 32: 314-317.
24. Wildung, R. E., K. M. McFadden, and T. R. Garland. 1979. Technetium sources and behavior in the environment. J. Environ. Qual. 8:156-161.

Relationship Between Land-Use Practices and Fecal Bacteria in Soils[1]

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ABSTRACT

Fecal and other bacteria populations in soils from pasture, cornfield, and forest sites were assessed at the Rhode River watershed. Total coliform (TC), fecal coliform (FC), total streptococci (TS), fecal streptococci (FS), and aerobic heterotrophic bacteria (TVC) numbers were determined seasonally. They were also determined with depth to 25 cm in soils from the pasture. Bacterial numbers per gram of pasture soil from the surface 1 cm were: 252-1,603 TC, 10-396 FC, 317-2,270 TS, 163-1,330 FS, and 1.6-65.1 × 10^6 TVC. The numbers declined markedly with depth, except that TS and TVC numbers remained near the surface values. Fecal coliform numbers varied seasonally, from 22 to 396 MPN/g soil in fall and spring when cattle were grazing, and from 10 to 50 MPN/g soil in winter and summer when cattle were absent from the pasture. The cornfield and forest surface soils contained equally low FC numbers ranging from 3 to 27 MPN/g soil. Aerobic heterotrophic bacteria were similar in magnitude at all three land-use sites. No readily discernable seasonal pattern was noted. The proportion of coliform bacteria differed with land use. In pasture soils 67% of isolates were confirmed as *Escherichia coli*, 22% were *Enterobacter aerogenes*, and 11% belonged to other species. In cornfield soils only 8% were *E. Coli*, 77% *E. aerogenes*, and 15% other species. In forest soils only *E. coli* were the dominant species. Thus, the proportion of *E. coli* was directly related to the degree of fecal pollution of the soils, and the proportion of *E. aerogenes* increased with remoteness of the soil from potential polluting agents.

Additional Index Words: **pasture, forest and cornfield soils, total and fecal coliforms, total and fecal streptococci, aerobic heterotrophic bacteria, soil discharge and retention.**

Faust, M. A. 1982. Relationship between land-use practices and fecal bacteria in soils. J. Environ. Qual. 11:141-146.

The Rhode River watershed is representative of the coastal plain of the eastern United States. The watershed has a mosaic of land uses, primarily row crops, pasture, and forested areas. As these areas are managed for a multitude of uses, water quality in the streams, and therefore the receiving estuary, is commonly affected. A comprehensive investigation of the fecal bacteria in water runoff from the Rhode River watershed into the estuary has so far indicated that the greatest concentrations of fecal bacteria originated from pasture land and the least from forested areas (Faust, 1976; Faust and Goff, 1977). There is, however, a lack of information as to whether fecal organisms are dislodged from the surface or move through the soils before entering the estuary through runoff.

Geldreich et al. (1962) investigated the relationship between land use and fecal coli and aerogenes bacteria in various soils and found that these organisms are present in relatively low numbers in undisturbed soils (i.e., "virgin forests") and that their numbers increased sharply in pasture soils. Fecal contamination of soil and the subsequent entry of contamination into water supplies presents a more complex situation than was noted from addition of diluted fecal material (Van Donsel et al., 1967; Evans and Owens, 1973). Soil types, the availability and survival rates of organisms during their residence in soil, and the likelihood of their being washed out by water runoff are factors to be considered. Since pollution of the soil surface presents a situation difficult to control at the source, knowledge of the characteristics of soil contamination is necessary. Therefore, this study was initiated to determine the concentrations of total and fecal coliforms and total and fecal streptococci in soils and to attempt to relate their characteristics to land-use practices and the possibility and distribution of estuarine contamination.

MATERIALS AND METHODS

Description of the Watershed

The Rhode River watershed is located on the western shore of the Chesapeake Bay, south of Annapolis, Md., and has a total watershed area of 3,332 ha. The watershed is divided naturally into experimental basins, which vary in size from 6 to 1,229 ha and are in land use from single- to multi-purpose use (Correll et al., 1978). The present study reports on only small, single land-use forest or mixed cornfield (*Zea mays* L.) and pasture partially covered with forested watersheds. These areas are as follows: a cornfield of 16.4 ha (No. 109), a forest of 6.3 ha (No. 110), and a pasture of 5.4 ha (No. 111) watersheds. Ten

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