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EROSIONAL BEHAVIOR OF CESIUM-137*

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Abstract—Movement of ^{137}Cs by runoff, erosion, and infiltration on a silt-loam soil in Tennessee is reported. A summary of a 2-yr study is given, and a model for the nuclide erosional loss is suggested. It was found that, although an empirical parabolic equation adequately describes the radionuclide loss, an exponential model, based on the nuclide distribution in a soil profile, appears to give more general results. The results show that a 60% reduction in the radiation dose due to surface-deposited ^{137}Cs occurred during the first 7 months following the application. Most of the applied radiocesium was found in the 0- to 3-cm layer or had been eroded away. It appears that the specific loss of ^{137}Cs reflects the seasonal variation in the magnitude of the erosion index and the extent of vegetative cover for a particular location.

INTRODUCTION

SIGNIFICANT accidental or purposeful contamination of terrestrial and aquatic environments with radionuclides is a potential danger to man or to his environment. In particular, the release and accumulation of slowly decaying radionuclides such as ^{137}Cs may pose a long-term hazard. The study described here was undertaken to learn more about the behavior of surface-deposited ^{137}Cs under natural field conditions over an extended period of time. Cesium generally is very strongly sorbed by soils of the temperate regions containing illite.[1–3] Its movement downward into the profile by infiltration is rather limited.[4,5] Movement of ^{137}Cs along the surface may occur, however, as a result of erosion. Translocation to lower-lying areas of a field, and eventually to streams, rivers, and reservoirs may be considerable. This is particularly true when soil is susceptible to erosion or when washoff of deposited ^{137}Cs from vegetation during a major storm takes place.[6–8]

In this paper erosional losses are summarized and reduction in the initial radiation dose, profile distribution, and seasonal fluctuations are considered. A model for predicting ^{137}Cs losses is also suggested and discussed against the background of findings at the Oak Ridge National Laboratory, Oak Ridge, Tennessee. For interested readers, the characteristics of this study area and preliminary findings are to be found in a report by ROGOWSKI and TAMURA.[6] Although the radionuclide of interest here is ^{137}Cs, it is felt that similar concepts may apply generally to other pollutants, particularly if they are subject to loss by runoff and erosion.

MEASUREMENTS

The studies were conducted on three 2.5- × 2.5-m plots of alluvial Captina silt loam soil on the U.S. Atomic Energy Commission's reservation at Oak Ridge (Fig. 1). A more detailed description of the soil and plots was given in earlier publications by ROGOWSKI and TAMURA.[6–8] In the fall of 1964, five millicuries of ^{137}Cs were applied in the form of a dilute spray to three plots representing three different cover conditions: Plot 1, bare soil; Plot 2, poor meadow; and Plot 3, good meadow. In the following 2 yr, runoff and erosional losses were measured. Radiation air dose measurements were made with a GM survey meter during the first 7 months after the application.

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REPOSITORY OAK RIDGE ENERGY SYSTEMS
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Human Subjects Project

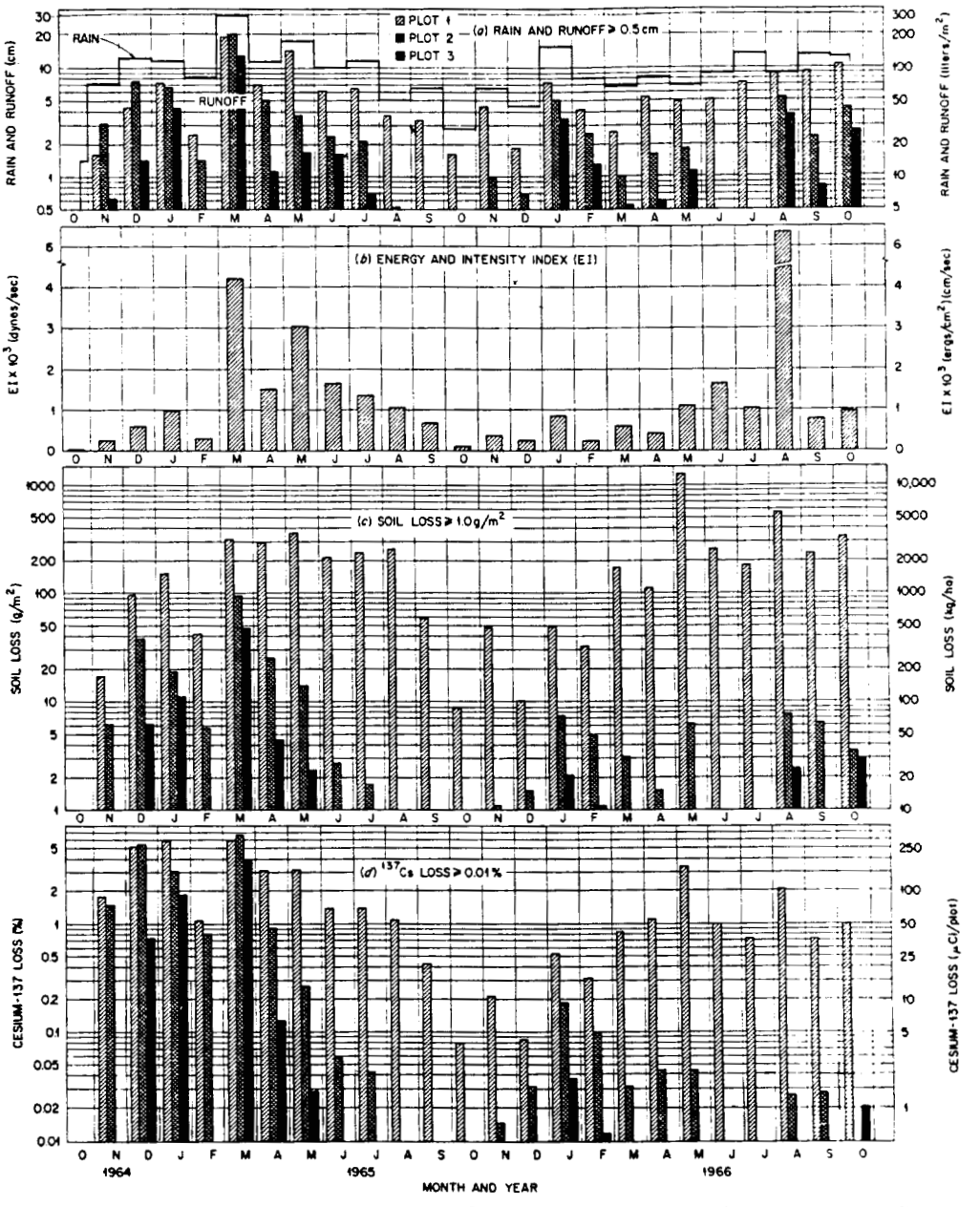

FIG. 2. Monthly rainfall and runoff ≥ 1.0 l./m² (a), erosion energy and intensity index (EI) (b), soil loss ≥ 1.0 g/m² (c), and cesium-137 loss $\geq 0.01\%$ (d), on three experimental plots Plot 1—bare soil, Plot 2—poor meadow, Plot 3—good meadow) on alluvial Captina silt loam soil, where 2.54 cm = 1 in., 0.4728 dyne/sec = 1 EI (ft-tons/acre) (in./hr), 224.2 g/m² = 1 ton/acre.

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FIG. 1. General view of radionuclide movement study plots located at the Oak Ridge AEC Reservation in Tennessee on the alluvial Captina silt loam soil.

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RESULTS

Soil and ^{137}Cs losses

Monthly values or rainfall, runoff, soil loss, and ^{137}Cs loss are presented in Fig. 2, and the 2-yr totals are given in Table 1.

Despite the small size of the study plots, soil loss, S, from these experimental areas followed quite well the empirical relationship derived by WISCHMEIER and SMITH:[10]

$$S = K \times \mathrm{EI} \times C \times \Lambda \qquad (1)$$

where

S = soil loss (g/m^2);

K = soil erodibility constant (g/m^2)/(dyne/sec), equal to 1.96 ± 0.19* for Captina soil;

EI = erosion index (dyne/sec), defined as the product of rainfall energy and its maximum 30-min intensity;

C = dimensionless cover correction factor, equal to 1.0, 0.04, and 0.006 for Plot, 1, 2, and 3 respectively;[7]

Λ = dimensionless topographic slope—slope-length correction factor, equal to 0.08, 0.12, and 0.26 for Plot 1, 2, and 3 respectively.[7]

Using the above numerical values, the predicted soil losses in column 6 of Table 1 were computed. Soil loss may also be expressed as an average thickness, D (cm), eroded:

$$D = S/(D_b \times 10^4) \qquad (2)$$

where

D_b is the average surface soil bulk density (g/cm^3) and 10^4 is the unit conversion factor (from m^2 to cm^2).

The total EI value of 30.3×10^3 dynes/sec for the 2 yr of experimentation was higher than expected and somewhat different in distribution than normal for the area (Fig. 3).

It was concluded that considerable erosional losses of ^{137}Cs can and do occur, particularly on exposed areas (bare soil—Plot 1). The highest losses from the vegetation-covered plots were observed during the first 7 months (Fig. 2), probably due to washoff from vegetation.[9] The results also indicated that uptake by new vegetation was minimal (less than 1% of the applied ^{137}Cs) and consistent with the findings of others.[11]

In Fig. 4, the relationship between cumulative loss of ^{137}Cs and loss of soil is shown for the bare soil (Plot 1):

$$Y_s = (A(S/b)^n \qquad (3)$$

where

Y_s is the cumulative ^{137}Cs loss as a percentage of the amount applied,

S is the cumulative soil loss (g/m^2),

b is a units constant = 1.0 g/m^2,

n is the exponent ($0 \leq n \leq 1$), and

A is a coefficient.

The coefficient A and exponent n were determined from the least squares fit of the logarithmic experimental data obtained after 153 days ($S \geq 800$ g/m^2). The curve (Fig. 4) was then extrapolated back to the origin. The estimated standard error* of Y_s was found in an extrapolated fit to be ±0.29%, and the greatest

* Estimated standard error,

$$s = \sqrt{(1/n)\sum_{i=1}^{n}(y_i - \bar{y})^2/(n-1)}; \quad n = 50.$$

Table 1. Two year total rain, runoff, infiltrate, soil, and cesium-137 loss on three study plots of alluvial Captina silt loam soil in Tennessee*

Plot	Rain (cm)	Runoff (cm)	Infiltrate (cm)	Soil loss (g/m^2)†		^{137}Cs loss (%)‡	
				Observed	Predicted	Soil	Solution
1	245	149	96	5258	4751	42.3	0.6
2	245	78	167	250	285	18.8	0.6
3	245	40	205	85	93	6.4	0.4

* Plot 1, bare soil; Plot 2, poor meadow; Plot 3, good meadow.

† Conversion factor is: 224.2 g/m^2 = 1 ton/acre; observed = experimentally observed; predicted = predicted from equation (1).

‡ Cesium-137 loss as a per cent of total applied (903, 937, and 925 μCi/m^2 of cesium-137 on Plot 1, 2, and 3, respectively, on October 20, 1964).

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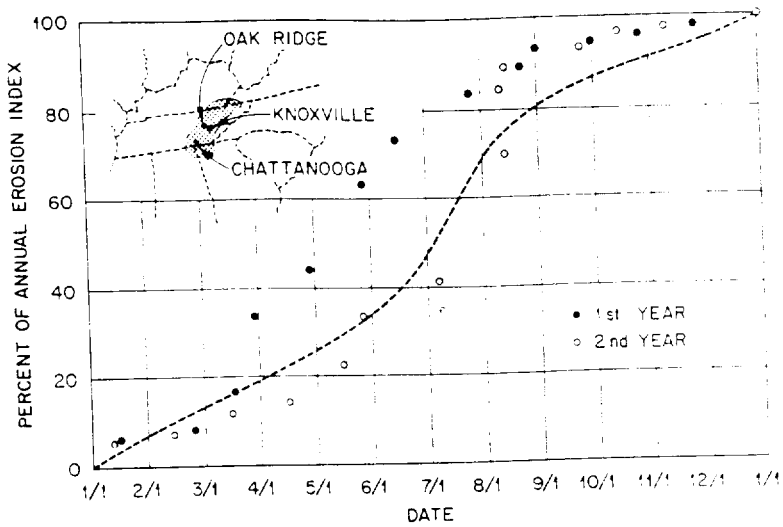

FIG. 3. Erosion index distribution for parts of Tennessee [Fig. 21 of Wischmeier and Smith (1965)] = dashed line, and observed values = points.

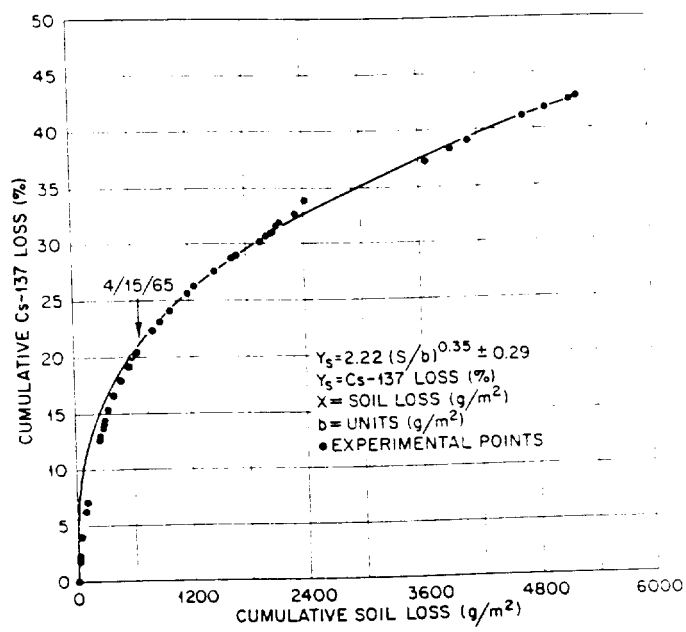

FIG. 4. Cumulative ^{137}Cs loss (%) as a function of soil loss (g/m²) on Plot 1 (bare soil). Final least squares fit for time greater than 153 days after application, fitted to entire data.

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deviations were in the lower range, where the fit overestimated the initial ^{137}Cs loss. Similar relationships, with different values of coefficients and exponents, could be applied to the meadow-covered plots, but, again, when developed on the basis of data obtained after 153 days, the equations overestimated the initial ^{137}Cs loss. The best least squares fits of equation (3) was obtained when the experimental data were split into three distinct time phases.[7]

RADIATION DOSE

Air dose rate measurements were made with a GM-survey meter following the application of ^{137}Cs in the fall of 1964. The details of the procedure are described in an ORNL Technical Memorandum by ROGOWSKI and TAMURA.[9] Survey of the data showed that the air dose rate measurements can be grouped into four time periods: 1 week, 6 weeks, 3 months, and 7 months, after the application: After 7 months the measurements were discontinued.

In Table 2, the average dose rate readings for the time intervals are given. The dose rate readings were taken with a calibrated GM survey meter with the window open. Actual ^{137}Cs losses in terms of percent of the amount applied are also shown for comparison. The highest initial dose rate (1 week reading) was observed on the poor meadow (Plot 2), the lowest on the good meadow (Plot 3). The ^{137}Cs applications in microcuries per square meter were 903, 937, and 925 for plots 1, 2, and 3, respectively, giving a ratio of 1.00:1.04:1.02; whereas the ratio of the initial dose rate readings was 1.00:1.07:0.98. It is possible, however, that the ^{137}Cs solution infiltrated into the soil on Plot 1 during the application, providing more initial shielding; it is also possible that the heavy vegetation on Plot 3 provided partial beta shielding. On the other hand, litter cover on the clipped meadow (Plot 2) would tend to intercept all of the tag spray at the ground surface, resulting in the highest initial dose rate reading.

In Table 3 the percentages in reduction of air dose rate measurements are shown. Little or no reduction in the radiation field was observed following the first rain; thus, the first week's measurements are here taken as a base reading of 100%. November rains, about equal in amount to October rains, but higher in regard to EI index, brought about the first reduction in dose rate measurements, and high EI rains in December 1964 and the early part of January 1965 produced another marked reduction. The largest and fastest reduction occurred on bare soil (Plot 1) during the first three months. In the last four months the reduction in dose rate was less than observed on the meadow plots. On bare soil (Plot 1), active erosion processes would tend to provide shielding in terms of covering up the radioactive material initially at the fastest rate, since all of the ^{137}Cs was at the ground surface. Therefore rapid initial reduction in rate would be

*Table 2. Average dose rate readings and cesium-137 losses as per cent of total applied on the bare soil Plot 1; poor meadow, Plot 2; and good meadow, Plot 3**

Time period	Cumulative time	Plot 1		Plot 2		Plot 3	
		Dose rate (mR/hr†)	^{137}Cs Loss (%)‡	Dose rate (mR/hr)	^{137}Cs Loss (%)	Dose rate (mR/hr)	^{137}Cs Loss (%)
October 20–28	1 week	2.68	0	2.89	0	2.63	0
October 28–November 30	6 weeks	1.87	1.8	2.47	1.5	2.29	0.004
November 30–January 12	3 months	1.11	10.9	1.53	8.5	1.75	2.7
January 12–May 12	7 months	0.93	11.3	1.12	8.6	1.29	4.0

* Original application of ^{137}Cs was at the rate of 903, 937, and 925 $\mu Ci/m^2$ on plots 1, 2, and 3, respectively, giving a ratio of 1.00:1.04:1.02.

† Dose rate reading in mR/hr at 1 m above ground.

‡ Cesium-137 losses between indicated times as per cent of total applied.

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Table 3. Radiation level given as an average percentage of first week readings on bare soil plot; poor meadow, Plot 2; and good meadow, Plot 3.
(Cumulative time is shown, and integrated rain amounts, EI values, and soil loss for the corresponding time intervals are listed.)*

	Rain (cm)	EI (dyne/sec)	Cumulative time	Per cent of initial dose rate		
				Plot 1 (%)	Plot 2 (%)	Plot 3 (%)
October 20–28	4.0	101	1 week	100.0	100.0	100.0
Soil loss (g/m²)				0	0	0
October 28–November 30	4.3	173	6 weeks	69.8	85.5	87.1
Soil loss (g/m²)				17	6	0.2
November 30–January 12	20.3	1496	3 months	41.4	52.9	66.5
Soil loss (g/m²)				242	57	17
January 12–May 12	55.1	7237	7 months	34.7	38.8	49.0
Soil loss (g/m²)				748	131	52

* Rain, EI, and soil loss values given for periods between indicated dates; 5 mCi of ^{137}Cs was applied to each plot on October 20, 1964.

expected and subsequent reduction should more nearly reflect the actual erosional losses. Thus a 7% reduction in dose rate on the Plot 1 in the last four months (Table 3) was accompanied by an 11% erosional loss (Table 2) of ^{137}Cs.

On meadow plots a different situation prevailed. Following the spray application all of the ^{137}Cs was initially deposited on vegetation. In the first six weeks 55% washoff from the vegetation occurred, in three months 65% was gone, by Spring (153 days) 72%, and by Fall (346 days) on the average 93% has been leached off. The reduction in dose on the meadow plots was also highest during the first three months decreasing with the amount of vegetation, although the rate of reduction was less than on the bare soil plot. In the following four months the reduction rate on both meadow plots remained approximately equal to the washoff rate but greater than the rate on bare soil—Plot 1. Retarded washoff from vegetation would continue adding ^{137}Cs at the soil surface where, because of the vegetative cover, the erosive processes would not be as active in covering it up as on the bare soil plot. This coupled with lower overall erosive losses, again reflecting the influence of vegetation, would result in lower initial reduction in the dose rate. Since the erosive processes are not as active on meadow plots as they are on the bare soil and since the ^{137}Cs is continuously being added through washoff at the surface relatively more of the active material is usually available for transport off the plot by erosion. This could explain a higher rate of dose reduction on meadow plots during the last four months especially so since by early Spring old dead vegetation made these plots less resistant to soil and nuclide losses by erosion. The details of the ^{137}Cs distribution within the plots as well as further data and discussion on leaching off the vegetation are presented elsewhere.[7] On the average, during the first 7 months of study under East Tennessee conditions on Captina silt loam soil, approximately a 60% reduction of the original dose rate was observed.

DISTRIBUTION OF ^{137}Cs IN SOIL PROFILE

The distribution of ^{137}Cs within the plots has been discussed in another publication by Rogowski and Tamura.[7] It was reported that the highest concentration of ^{137}Cs tended to accumulate in the depressions close to the exits of the plots' runoff. In an effort to obtain a relative distribution in the soil, sample cores were taken, sliced into horizontal sections, and analyzed. The ^{137}Cs content in each section was computed as a fraction of the total in the profile at the sampling point.

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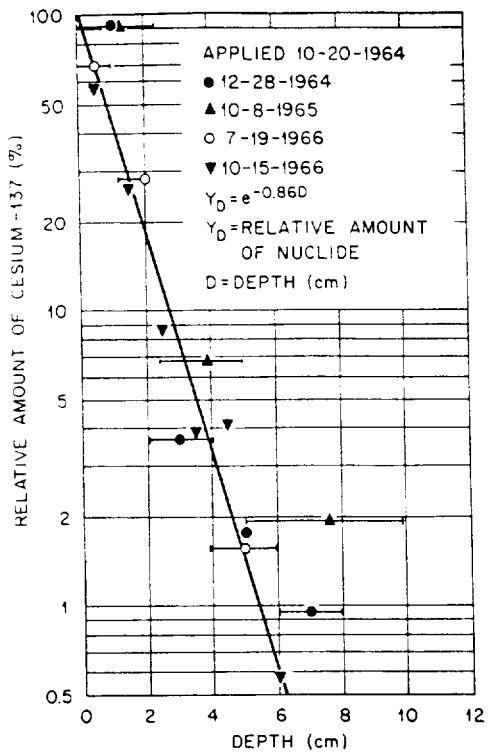

FIG. 5. Relative distribution of ^{137}Cs with depth on Captina silt loam.

The average results of all three plots at different sampling times are presented in Fig. 5. The data points for October 15, 1966, are an average of 72 observations as contrasted with a total of 17 observations for other points. The equation forced to pass through (0, 100) and fitted to the October 15, 1966, data points has the following general form:

$$Y_D = e^{-\lambda D} \tag{4}$$

where

Y_D is the fraction of ^{137}Cs present in the profile below depth of D centimeters and
λ is a constant (equal to 0.86/cm).

More than $\frac{9}{10}$ of the ^{137}Cs present in the profile was in the upper 3 cm of the soil. More than half of this fraction appeared to be near the surface where it was most susceptible to loss by runoff and erosion.

SEASONAL FLUCTUATIONS

The time dependence of ^{137}Cs loss by erosion was not apparent in the results presented earlier.[6] Subsequent analyses suggested that the interaction between radionuclide loss and soil is dependent on time. The nature of this time-dependent relation is shown in Fig. 6. Specific loss of ^{137}Cs, defined here as nuclide loss per unit weight of soil per unit of erosion index (EI), is plotted on a logarithmic scale as a function of time after the application of ^{137}Cs in the fall of 1964. A general least squares fit of a straight line to the data from all the plots and the 95% confidence limits are also shown. The resulting equation for Captina soil under East Tennessee conditions may be written:

$$Y_t = 5.1 \times 10^{-3} e^{-8.2 \times 10^{-3} t} \tag{5}$$

where

Y_t = specific loss of ^{137}Cs [(μCi/g)/(dyne/sec)],
t = time (days).

Equation (5) approximates the specific loss of ^{137}Cs at a particular time following application; it should not be used for computation of the cumulative loss, since both the soil loss and EI parameters are in themselves time-dependent functions of rainfall distribution. It is evident that large seasonal deviations from the mean take place. The general downward slope of equation (5), however, is indicative of surface depletion of the radionuclide, both by runoff and erosion and by movement into the soil profile. Figure 6 shows the extent of the seasonal fluctuation of specific ^{137}Cs loss. In the late fall, winter, and early spring, the specific loss is generally higher than the mean value given in equation (5); for the rest of the year it is generally lower. For an explanation, one has to consider the differences that exist between winter and summer storms in Tennessee (Table 4). In winter, low EI storms are common. Under these conditions, mostly fine clay material would have been eroded from the experimental plots. Since ^{137}Cs is adsorbed on the clay minerals of soil, high specific loss values would result. In contrast, summer rains come in the form of short, high EI thunderstorms. These storms are capable of eroding larger quantities and coarser soil material, thereby "diluting" the cesium-carrying clay fractions. The curve in Fig. 6 represents an overall seasonal trend for all plots lumped together. The inferred conclusion that the magnitude

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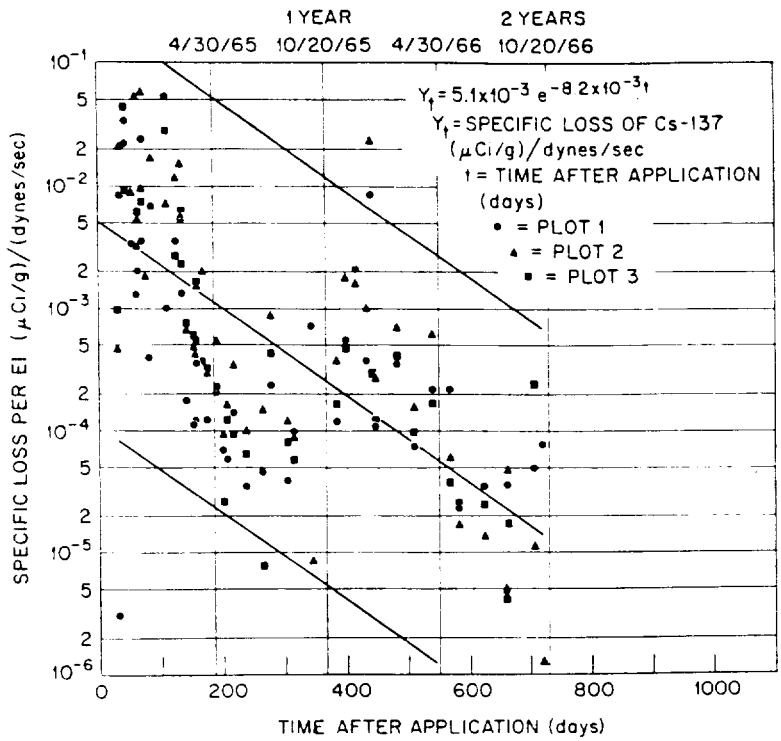

FIG. 6. Seasonal distribution of ^{137}Cs loss per unit EI where EI is given in dynes/sec.

of the EI index is responsible for variation seems justifiable, since the protection against erosion afforded by the vegetation tends to be offset by greater availability of ^{137}Cs at the soil surface of meadow plots as discussed earlier. However the protective effect of actively growing vegetation in the summer should not be discounted since by limiting erosive losses it could also lower the specific loss values.

In the discussion above, the specific cesium losses given in Fig. 6 should not be identified with the total amounts lost. The results presented in Fig. 7 show that total losses do not necessarily correspond to specific losses. All the plots showed highest losses during the first winter. During the second year, however, a higher loss was observed on the bare soil (Plot 1) during the spring and summer than during the winter. The opposite was true for the meadow plots (2 and 3). The contrasting behavior of the bare soil and meadow plots can be explained by considering the erosive properties of the soil. Summer thunderstorms, having high energy and intensity, can erode

Table 4. Seasonal comparison of the average number of rains; erosion index, EI; and EI per rain for the 2-yr study

Date	Average number of rains	EI (dyne/sec)	EI per rain (dyne/sec)
October 10–December 22	10	476	48
December 22–March 21	15	2086	146
March 21–June 22	24	5278	216
June 22–October 20	28	7301	257

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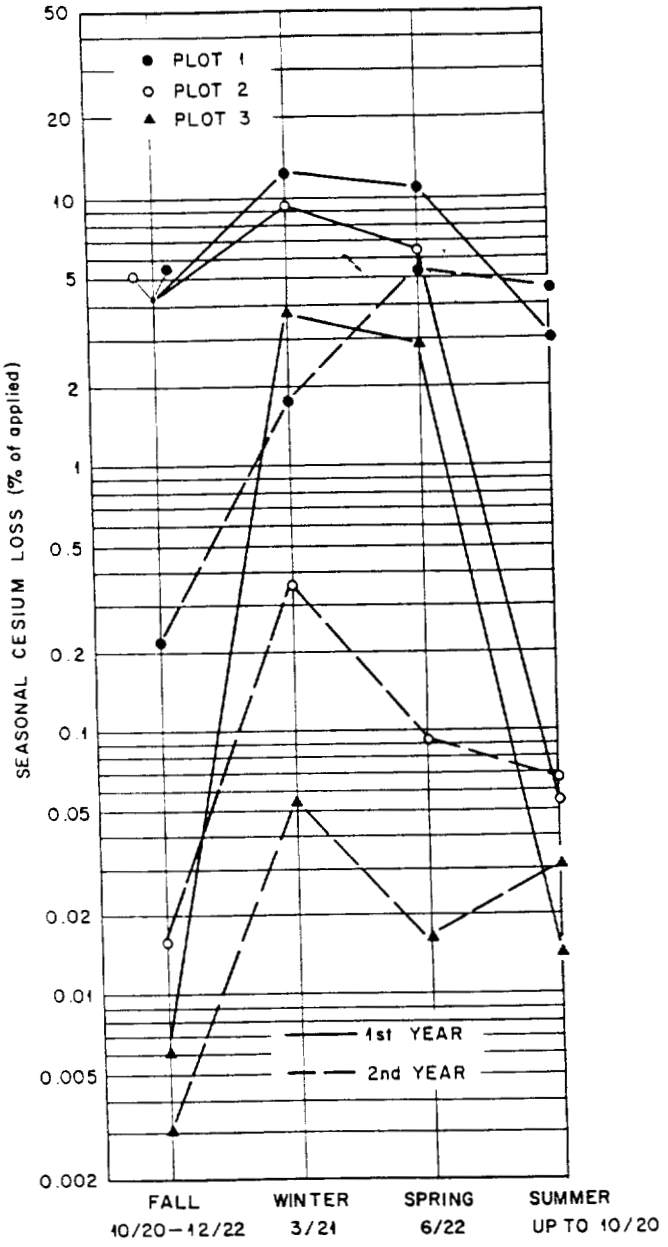

FIG. 7. Seasonal distribution of ^{137}Cs loss on bare soil (Plot 1): Poor meadow (Plot 2), and good meadow (Plot 3) for the 2 years of study; October 20, 1964, was the application date.

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a considerable quantity of soil from exposed bare soil areas such as Plot 1 after a surface seal and crust develop. Under meadow use, the actively growing vegetation depletes the moisture in the profile and permits more storage of water from summer storms. In addition, the surface layer of organic matter enhances infiltration, and the vigorously growing meadow provides adequate protection for the surface soil. Thus, it is during the winter months, when the profile is saturated and the vegetation is dead, that one might expect both the soil and ^{137}Cs losses on the meadow plots to be largest, such as Plots 2 and 3.

EROSIONAL LOSS MODEL

The results discussed above indicate that a relationship [equation (3)] exists between ^{137}Cs loss (Y_s) and soil loss and that, although most of the ^{137}Cs is in the upper centimeter of the soil [equation (4)], reduction in radiation dose rate near the surface in the first 7 months after application amounts to 60 % of the original dose rate. Specific ^{137}Cs losses, Y_t, appear to depend on seasonal fluctuations in erosion index [equation (5)], and the profile distribution, Y_D, is of an exponential type [equation (4)].

With this information as background, one may proceed to formulate a general model of erosional losses of radiocesium. It was stated previously that equation (4) is based on core scan data. The value of Y_D estimates the amount of nuclide present below a given depth, D, as a fraction of the total in a profile. Thus the fractional amount present in the upper section of thickness D is

$$Y_\lambda = 1 - e^{-\lambda D}, \tag{6}$$

and

$$Y_\lambda + Y_D = 1. \tag{7}$$

Solving for λ,

$$-\lambda = \frac{\ln(1 - Y_\lambda)}{D}. \tag{8}$$

Supposing D is taken as a thickness of soil eroded, then equation (6) should give the amount of nuclide loss as a fraction of the total present in the profile. If the nuclide distribution after 2 yr of observation is indeed given by equation (4), then the values of λ for some final period as calculated from equation (8) should be numerically equal to λ in equation (4), obtained from the core scan data.

In Table 5 the values of λ_1, computed on the basis of the last 2½ months' data (D_1, Y_{D_1}),* are indeed approximately equal to the value of λ in equation (4). By a similar argument, the values of λ_2, computed relative to the total ^{137}Cs applied originally, may be obtained (Table 5).

It is assumed that the values of λ_1 can be considered to be approximately the same for all plots. For bare soil (Plot 1) $\lambda_2 \sim 2\lambda_1$, the values of λ_2 for the meadow plots are about eight times the value of λ_2 for Plot 1. The discussion above thus suggests the following model of nuclide loss by runoff and erosion;

$$Y_\lambda = 1 - e^{-2n\lambda_1 D}. \tag{9}$$

Here λ_1 refers to some experimental or hypothetical distribution of nuclide in a profile. The expected thickness of layer eroded, D, may be computed from equation (2). The exponent n takes on different integral values ≥ 1, depending on whether the nuclide was initially deposited on the vegetation or directly on the soil surface.

In Table 6, losses of ^{137}Cs estimated with the erosion index probability data of WISCHMEIER

* Abstracted from Appendix 7 of Ref. 9.

Table 5. Depth eroded, D; per cent of ^{137}Cs lost, Y_D; and computed values of exponent λ; relative to final 2-½-month ^{137}Cs loss (subscript 1) and total originally applied ^{137}Cs loss (subscript 2)

Plot	$\times 10^{-4}$ cm		Per cent		λ_1	λ_2
	D_1	D_2	Y_{D_1}	Y_{D_2}		
1	373	3482	2.96	42.86	0.81	1.61
2	6.50	166	0.045	19.41	0.69	12.99
3	2.69	57	0.027	6.83	1.00	12.42

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Table 6. Computed values of ^{137}Cs *loss** Y_λ *under bare soil and good meadows conditions using erosion index values with different probabilities or occurrences†*

	EI‡ ($\times 10^3$ dynes/sec)	Bare soil§		Good meadow‖	
		D (cm)	Y_λ (%)	D ($\times 10^{-2}$ cm)	Y_λ (%)
Computed for minimum EI	15.4	0.16	24.1	0.21	2.8
Computed for maximum EI	44.3	0.46	54.6	0.60	7.9
Observed values	30.3	0.35	42.3	0.56	6.4
Computed for EI having 25% probability (P_2)	25.4	0.26	36.5	0.34	4.6
Computed for EI having 4% probability (P_2)	32.9	0.34	44.4	0.44	5.9

* Computed from equation (9) with $\lambda_1 = 0.86$ and $n = 1$ and 8 for bare soil and good meadow respectively.

† Values abstracted from 50 and 20% probability columns in Table 11 of Wischmeier and Smith, 1965, for Chattanooga, Tennessee. The values listed in above reference are for probability P_1 for a single year. Probability P_2 that consecutive years will have double that value is given by $P_2 = P_1^2$.

‡ The values of EI are for Chattanooga, Tennessee, since normal precipitation at Oak Ridge is more like that of Chattanooga rather than Knoxville. Both Chattanooga and Knoxville fall within an area of EI distribution that also contains Oak Ridge (Fig. 2). Minimum and maximum EI values given in Wischmeier and Smith, 1965, have the units 0.01 ft-ton/acres in./hr to obtain values for 2 yr (column 1, rows 1 and 2); they were multiplied by 2 and by 47.28 (dyne/sec)/(0.01 ft-ton/acre)(in./hr); they represent 22 yr range.

§ Thickness of the layer eroded on bare soil plot was computed using $A = 0.08, C = 1, K = 1.96, D_b = 1.51$ in equations (1) and (2).

‖ Thickness of layer eroded on good meadow plot was computed using $A = 0.26\ C = 0.004$ (standard value of good meadow cover correction factor C is different from observed 0.006), $K = 1.96$, and $D_b = 1.51$.

and SMITH[10] and equations (1), (2) and (9) are given for the 2-yr study. The observed results are also listed for comparison. The model appears to give satisfactory results on both the bare and good meadow plots. Observed erosion index value for the 2-yr study was considerably higher than expected normal (19.8×10^3 dynes/sec)[7] for Oak Ridge and appears to be close to 4% probability value (Table 6). In the probabilistic sense it may have a recurrence interval of about 25 yr or approximately equal to the half-life of radiocesium.

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