UNIVERSITY OF WASHINGTON

LABORATORY OF RADIATION BIOLOGY

SEATTLE, WASHINGTON 98105

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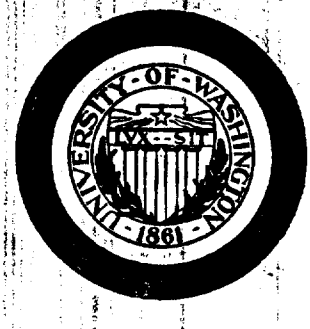

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ATOLL SOIL TYPES IN RELATION TO THE DISTRIBUTION OF FALLOUT RADIONUCLIDES

E.E. Held, S.P. Gessel and R.B. Walker

Laboratory of Radiation Biology
University of Washington
Seattle, Washington

Lauren R. Donaldson
Director

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ABSTRACT

The redistribution of radionuclides in atoll soils following fallout from a nuclear device is described. The soils are calcareous, containing no inorganic colloids, and their exchange capacity is directly related to organic content. Comparison of gamma-ray spectra of depth increments from young and old soils shows that Cs^{137} and Sb^{125} move most readily in old soil, while the principal gamma-emitting radionuclide moving in young soil is Sb^{125}. Sr^{90} moves in both old and new soils, and quantitative differences in vertical movement between soil types is obscured by the highly variable surface distribution of the radionuclides. There is a vertical gradient in the distribution of radionuclides even within the surface inch. Litter redeposits Cs^{137} and Sr^{90} at the soil surface and bird droppings have added Zn^{65} and Co^{60}. In young soils the highest levels of radioactivity are associated with soil algae found as a surface crust in undisturbed areas and in coral fragments in eroded areas. Horizontal movement is localized and probably is of little overall importance. Buried organic horizons contain more Cs^{137} than adjacent soil layers, and roots are generally more radioactive than the surrounding soil except at the soil surface. Pumice particles in the soil adsorb radionuclides but pumice is found infrequently. Mechanical mixing by animals in old soils and by erosion in young soils

is important in the redistribution of radionuclides near the soil surface. Cs^{137} and Sr^{90} are the principal radionuclides entering a cycle within the soil-plant system. Any loss from this system appears to be small, but a definite conclusion can not be drawn from the data.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

ATOLL SOIL TYPES IN RELATION TO THE DISTRIBUTION OF FALLOUT RADIONUCLIDES

INTRODUCTION

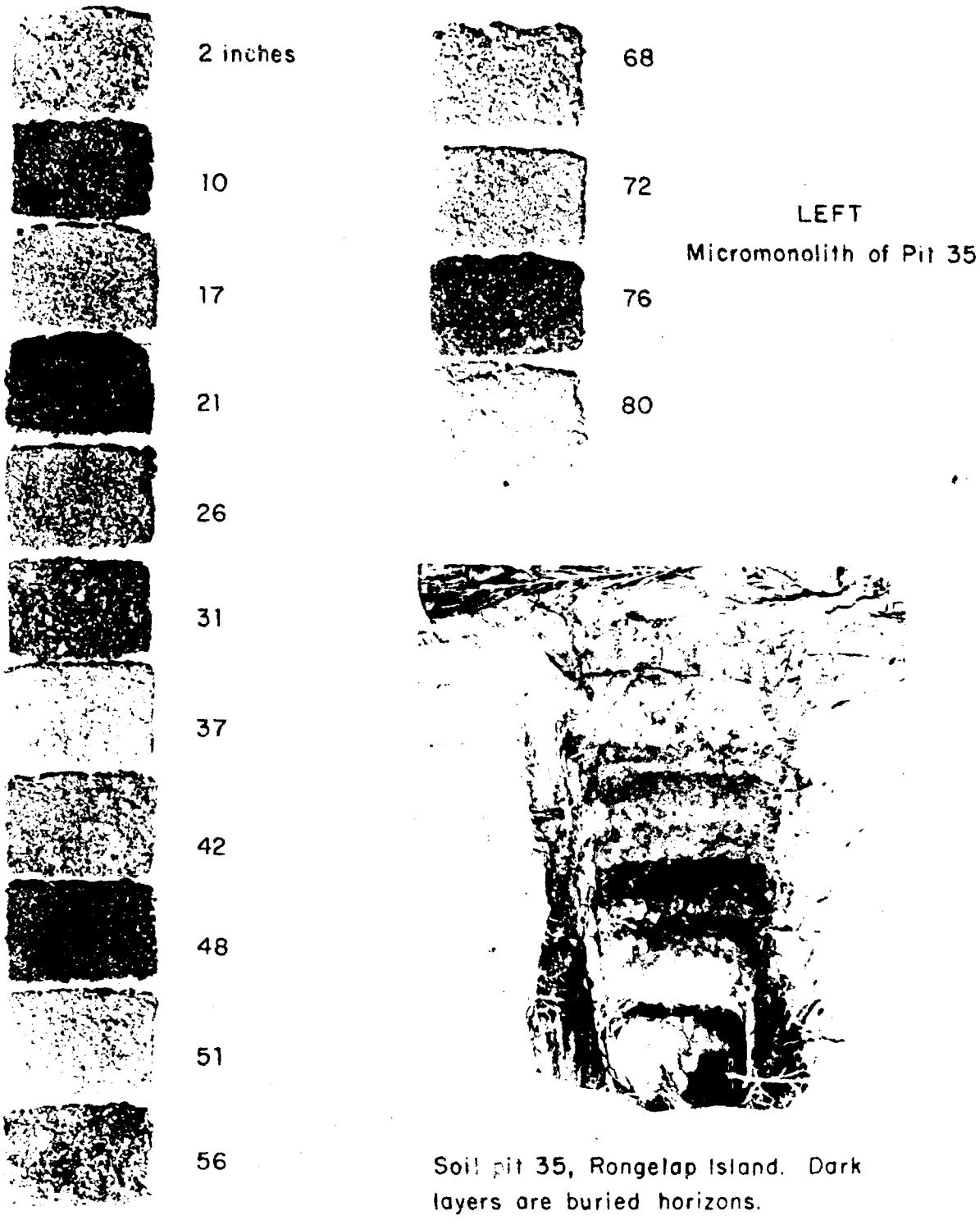
The redistribution of radionuclides in atoll soils following contamination with radioactive fallout is the subject of this paper. Rongelap Atoll, northern Marshall Islands, in the central Pacific Ocean, presents a unique opportunity for such studies since it was substantially contaminated with radioactive fallout only once. The fallout resulted from a thermonuclear device detonated at Bikini Atoll eighty miles to the west on March 1, 1954. Although there was some additional contamination from nuclear tests in 1956 and 1958 the total contribution of radionuclides from the fallout of these subsequent test series amounted to a fraction of one per cent of the amount from the 1954 fallout. Gamma radiation dose rates at Rongelap at detonation plus one day ranged from 3.5 r/hr at the southern islets of the atoll to 35 r/hr at the northern islets (Dunning 1957). These rates declined at approximately the rate predicted for mixed fission products by Miller and Loeb (1958).

Rongelap Atoll has a lagoon area of 388 square miles and an average depth of 168 feet (Nugent 1946, p. 748). The emergent land area is about three square miles, consisting of

sixty-one small islets ranging in size from a fraction of an acre to the largest, Rongelap, which is four miles long and one-half mile across at its widest point. There is one small islet on the western reef and the other islets extend along the northern, eastern and southern reefs. The islets on the northern reef are not as well developed as those to the east and south. There are two seasons--a dry season from December to March and a wet season from April to November. Annual rainfall is less than fifty inches, and there is no well-developed fresh-water lens. Some important features of Rongelap Atoll including aerial photographs are given by Wiens (1962).

Classification and mapping of the soil types at Rongelap Atoll were reported by Kenady (1962). The parent material is primarily calcium carbonate, originating from corals, foraminifera, coralline algae and mollusk shells. There is a very small amount of pumice drift in the soils. Since these soils contain no inorganic colloids, exchange capacity and organic content are linearly related. In some areas, particularly along the seaward sides of the islets, buried A_1 horizons are found as deep as eighty inches (Fig. 1). These highly organic horizons presumably result from storm debris covering previously established soil and vegetation.

The pH, determined in the field from a 1:1 soil-water ratio with a Beckman Model N-2 pH meter, is generally between



Soil pit 35, Rongelap Island. Dark layers are buried horizons.

Fig. 1. Beach Ridge Sand, Rongelap Island. This soil type occurs on the island beach ridges. The pit shown above is seven feet deep and contains six buried horizons.

7 and 8 in the surface horizon but occasional values from 4.6 to 9.5 were found. The pH increases with depth and decreasing organic content.

The amounts of exchangeable cations in the different soil types are given in Table I . The calcium content is so high that more calcium is brought into solution with repeated extractions. Consequently, strontium units have little or no significance relative to atoll soils and, when given, are based on total calcium rather than on exchangeable calcium.

METHODS OF COLLECTION AND MEASUREMENT

The vertical distribution of radioactivity in soil and litter was studied by analyzing samples taken mostly by 1-inch soil increments, in a few cases by 1/4- and 1/8-inch increments and by radioautographs of sections of soil cores prepared by the method of Held et al. (1965). The increment collections were made during both the wet and dry seasons in 1958, 1959, 1961 and 1963. The cores were collected only in 1963. Because there is considerable horizontal variation in the levels of radioactivity (Table II) each set of increments was collected to insure the sampling of a single vertical column.

The large amount of horizontal variability also made it more profitable to compare the relative amounts of radionuclides at different depths from many profiles than to make precise

Table I. Nutrient levels and soil reaction in the surface A_1 horizon of the five most extensive soil series (from Kenady 1962).

Soil Series	Rongelap Gravelly Sand	Gogan Gravelly Sandy Loam	Lomuilal Sand	Beach Ridge Sand	Kabelle Sand
Per Cent Nitrogen	0.57	1.71	0.26	0.09	0.14
Per Cent Organic Matter	16.7	35.6	6.4	4.5	7.7
*Exchange Capacity	22.2	37.7	12.6	3.7	5.7
*Sodium	3.36	4.01	2.68	1.16	1.52
*Magnesium	4.19	11.06	3.21	2.55	1.92
*Potassium	1.95	1.80	0.79	0.37	0.46
Phosphorus (ppm)	81.7	985.2	54.2	32.8	32.1
pH	8.1	7.8	8.4	8.6	8.6

* meq per 100 grams of oven dry soil

Table II. Sr^{90} levels in soil samples from two sets of plots. Samples collected in September 1961. Values in pCi/gm dry. Each plot is 16-1/2 by 33 feet.

Kabelle Islet, Pit 38 Area

Depth in inches	Plot		A		B		C		D		E		F		G	
	Sample		1	2	1	2	1	2	1	2	1	2	1	2	1	2
0-1	Subsample	1	100	110	94	81	220	280	530	82	40	69	71	100	76	62
	" "	2	78	100	95	94	240	200	410	130	49	81	49	100	91	73
1-2	" "	1	33	62	180	60	150	41	45	61	22	54	30	26	13	29
	" "	2	25	51	180	91	160	43	49	78	24	52	29	22	21	22
2-3	" "	1	44	50	33	50	65	29	41	49	8	36	19	14	8	17
	" "	2	36	52	25	69	79	23	41	51	11	32	25	17	7	21

Rongelap Islet, Pit 22 Area

Depth in inches	Plot		A		B		C		D		Pit 22	
	Sample		1	2	1	2	1	2	1	2	1	2
0-1	Subsample	1	230	230	130	110	180	170	160	180	130	140
	" "	2	230	150	150	170	190	180	150	350	150	200
1-2	" "	1	150	190	84	47	100	53	54	140	77	100
	" "	2	160	180	89	47	95	61	63	140	79	97
2-3	" "	1	63	49	19	6	28	13	25	65	25	43
	" "	2	55	50	18	4	30	12	21	65	19	36

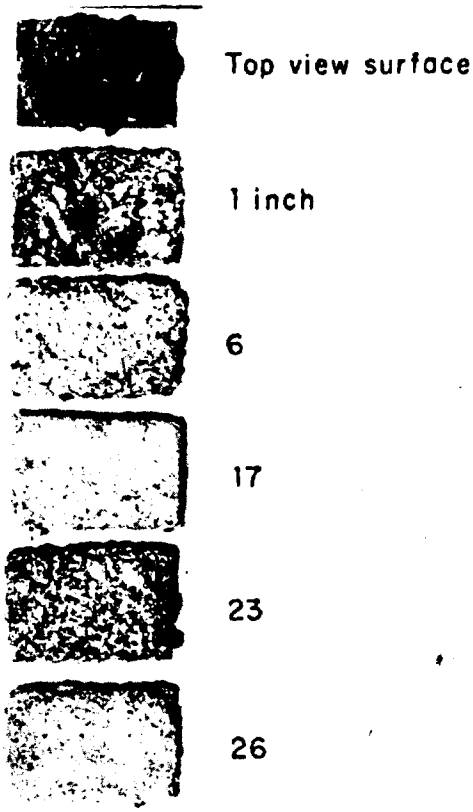
quantitative determinations from a few profiles. Gamma-ray spectra of equal amounts of soil from different depths were compared directly to arrive at (1) a qualitative evaluation of the vertical distribution of the radionuclides and (2) a semiquantitative estimate of relative amounts of radionuclides at different depths. Gamma-ray spectra were made with a system which included a 3-inch by 3-inch solid, thallium-activated, sodium iodide crystal and a 256-channel analyzer. In addition, selected samples were taken for analyses of the pure beta-emitter, Sr^{90}.

RESULTS AND DISCUSSION

Vertical distribution in relation to soil type is made by comparing young and old soils. Examples are Kabelle Sand, a young soil, (Fig. 2) and Gogan Gravelly Sandy Loam (Fig. 3) and Lomuilal Sand (Fig. 4), old soils. Their characteristics are given in Tables I and III.

The young soil has little organic material except that in an algal surface crust about 1 cm thick. Hermit crabs, Coenobita perlatus, at the base of a few shrubs, the borrowing ghost crab, Ocypode ceratopthalmia, and ants are the main animals present. Litter accumulation is found only at the bases of the scattered shrubs and is a minor part of this soil system.

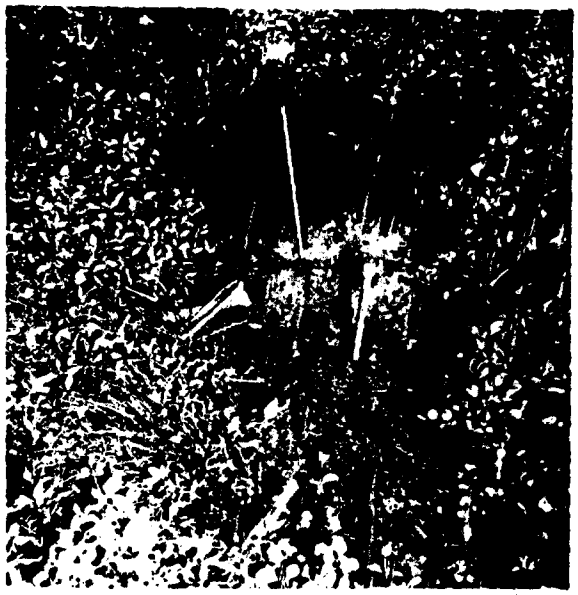
Soil pit 6. Black areas are crusts formed by algae.



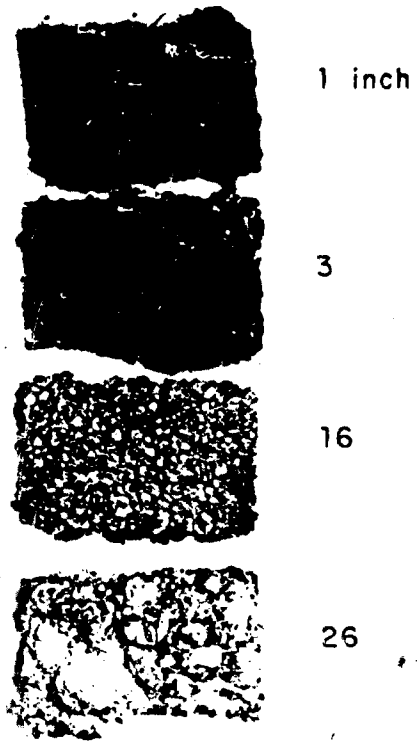
RIGHT: Micromonolith of pit 6.

General view of pit 6 area.

Fig. 2. Kabelle Sand. A very young soil composed of fine lagoon sands deposited on the lagoon side of Kabelle Island.



Pit 4, Gogan Series, Kabelle Island



Micromonolith of Pit 38

Stand of Pisonia grandis, Kabelle Island, pit 38 area.

Fig. 3. Gogan series, Kabelle Island. This is a well-developed, productive soil, usually associated with Pisonia grandis and Cordia subcordata.

Soil pit 12, Kabelle Island. Dark strata are buried horizons

Messerschmidia argentia at soil pit 12.

Micromonolith
Pit 21

Fig. 4. Lomuilal Sand, Kabelle Island.

Table III. Composition of representative soil types from Rongelap Atoll (after Kennedy 1962).

Kabelle Sand, a young soil

Soil Depth - inches	0-1	1-11	11-38
Per Cent Material > 2 mm	34	8	2
Per Cent Nitrogen	0.22	0.02	0.01
Per Cent Organic Matter	8.1	--	2.6
* Exchange Capacity	6.3	0.3	0.1
* Sodium	1.57	3.01	1.97
* Magnesium	3.37	1.04	1.16
* Calcium	3.01	2.88	2.65
* Potassium	0.57	0.15	0.20
Phosphorus (ppm)	30.0	12.0	12.0
pH	8.9	9.1	9.2

Lomulal Sand, an old soil

Soil Depth - inches	0-3	3-10	10-12	12-21	21-48
Per Cent Material > 2 mm	0	5	18	12	2
Per Cent Nitrogen	0.29	0.07	0.08	0.04	0.02
Per Cent Organic Matter	2.8	2.3	2.2	1.9	1.7
* Exchange Capacity	14.2	2.3	2.6	1.1	0.6
* Sodium	2.73	0.73	0.82	0.82	0.84
* Magnesium	4.66	8.36	1.05	0.85	0.83
* Calcium	5.02	1.87	2.50	2.31	2.35
* Potassium	1.09	0.18	0.19	0.16	0.16
Phosphorus (ppm)	105.9	15.1	14.1	5.0	5.0
pH	8.4	8.6	8.3	8.8	9.1

Gogan Gravelly Sandy Loam, an old soil

Soil Depth - inches	1-0	0-1	1-5	5-12	12-20	20-26
Per Cent Material > 2 mm	10	20	20	27	39	56
Per Cent Nitrogen	1.54	1.96	0.42	0.18	0.07	0.05
Per Cent Organic Matter	21.4	--	5.9	6.8	2.6	2.6
* Exchange Capacity	20.5	43.6	17.9	7.2	2.6	1.7
* Sodium	2.0	3.0	0.8	0.4	0.4	0.4
* Magnesium	7.0	7.4	4.0	2.2	1.2	1.1
* Calcium	10.3	24.2	14.1	6.2	7.7	7.8
* Potassium	--	1.80	--	--	--	--
Phosphorus (ppm)	1930.4	892.8	415.5	216.3	150.6	25.1
pH	7.4	7.1	7.9	8.2	8.6	8.8

* meq per 100 gm dry soil

In contrast, the old soil has well-developed A_0 and A_1 horizons and supports dense vegetation, which produces a heavy litter fall during the dry season and which contributes to the redistribution of radionuclides in the system.

Sea birds nest in the vegetation and land crabs, predominantly Birgus latro and Coenobita perlatus, burrow in the soil. Earthworms are seldom found and terrestrial isopods, although found in old soils on some islets, are few. There are also few soil insects, which are mainly tenebrionid and carabid beetles as well as ants.

Depth Gradient of Gamma Spectra

Old Soil

The gamma spectra, with background subtracted, of the 0 to 1-inch, 1 to 2-inch, and 9 to 10-inch depth increments from old soil collected in March 1959 are given in Fig. 5. The spectra of the increments between 2 inches and 9 inches have been omitted from the figure for clarity but show a gradual change from the condition at 1 to 2 inches to that at 9 to 10 inches. The radionuclides corresponding to the photopeaks are indicated in the figure. The photopeaks of Co^{60}, Zn^{65}, Mn^{54}, Ce^{144}-Pr^{144} and Eu^{155} show significant counts in the surface increments but are not detectable in samples from greater depths. The amounts of Cs^{137} and Sb^{125} decrease and the proportions of Cs^{137} and

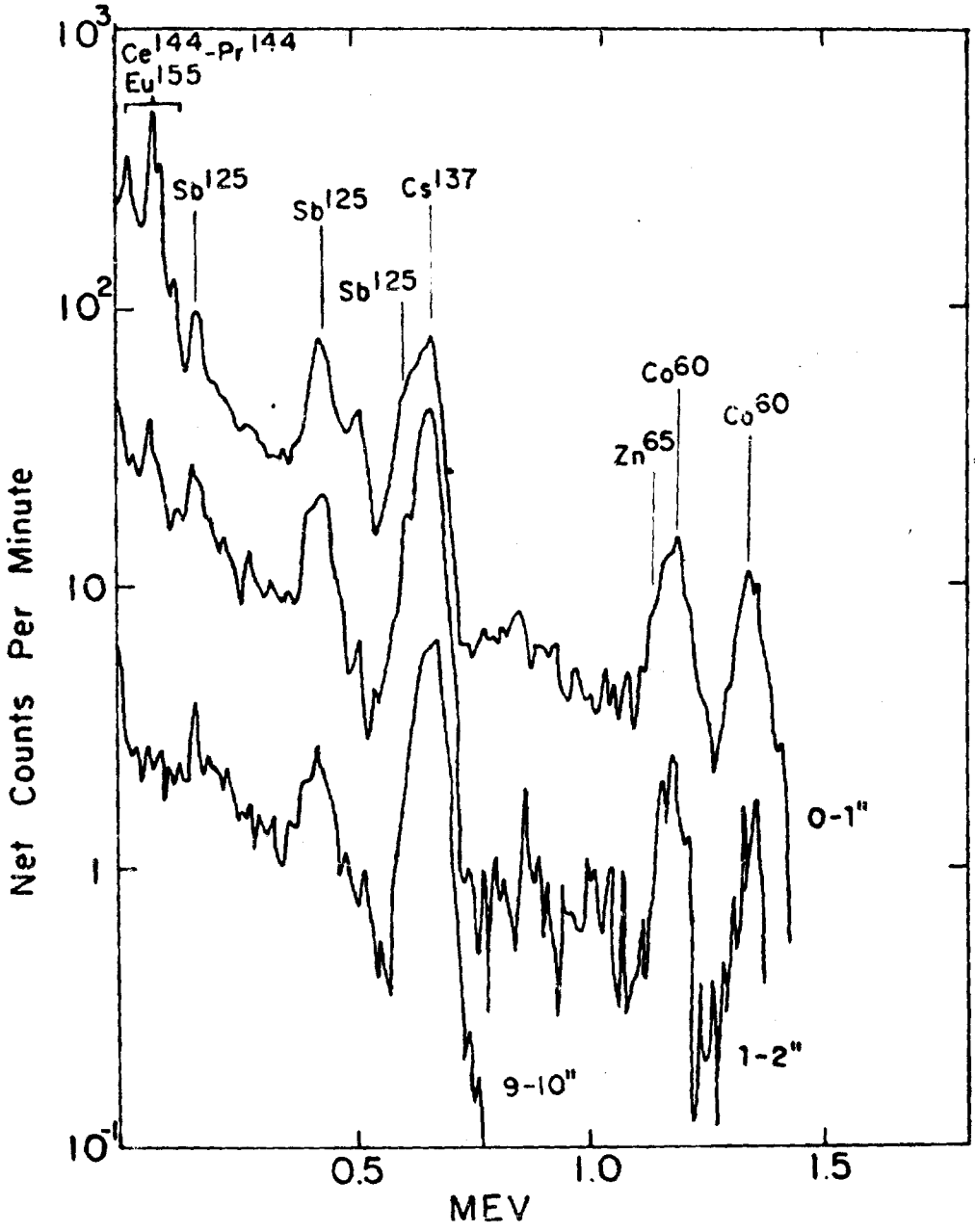


Fig. 5. Gamma-ray spectra of equal amounts of old soil from different depths.

Sb^{125} change with increasing depth. In the 0.60 to 0.66-mev photopeak region of the 0 to 1-inch increment, the relatively broad peak is a combination of the 0.60-mev photopeak of Sb^{125} and the 0.66-mev peak of Cs^{137}-Ba^{137m}. In the 1 to 2-inch increment the peak becomes sharper and is oriented toward the 0.66-mev photopeak of Cs^{137}-Ba^{137m}. At the 9 to 10-inch increment there is almost complete orientation toward the Cs^{137}-Ba^{137m} peak, with little Sb^{125} remaining. In Fig. 6 a comparison is given of the spectrum of the 9 to 10-inch increment (Fig. 5) and the gamma spectra of Cs^{137} and Sb^{125} spikes.

Fig. 7 shows the gamma spectra of increments taken from an undisturbed area in 1958. The first spectrum is from the 0 to 1/4-inch depth and the subsequent spectra are from 1/8-inch depth increments to a depth of 1 inch. The highest levels of Sb^{125}, the rare earths, and Co^{60}, which move more slowly than Cs^{137} or Sr^{90}, are in the 1/2 to 5/8-inch increments, whereas the Cs^{137}, which moves most rapidly in this soil type, is in the 3/4 to 7/8-inch increment.

Litter and Guano

The gamma-ray spectrum of litter, consisting of leaves, twigs and floral parts splattered with tern droppings, collected from old soil in 1961 (Fig. 8) shows the Cs^{137} photopeak to be much higher than the Sb^{125} peak. The 1.17 peak of Co^{60} is skewed to the left, indicating the presence of the 1.12-mev

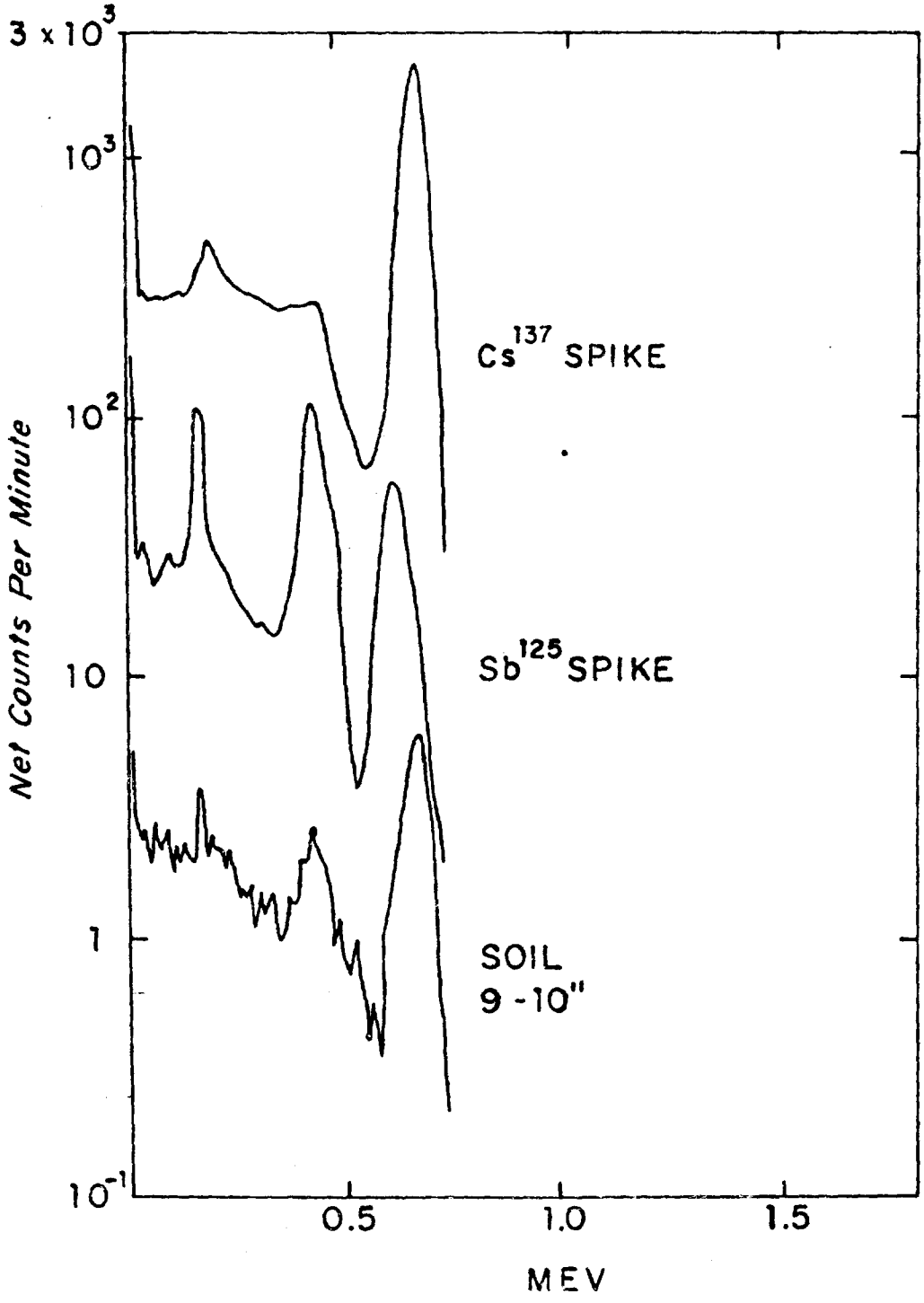


Fig. 6. Gamma-ray spectrum of a 9 to 10-inch depth increment from old soil compared with the gamma spectra of Sb^{125} and Cs^{137}.

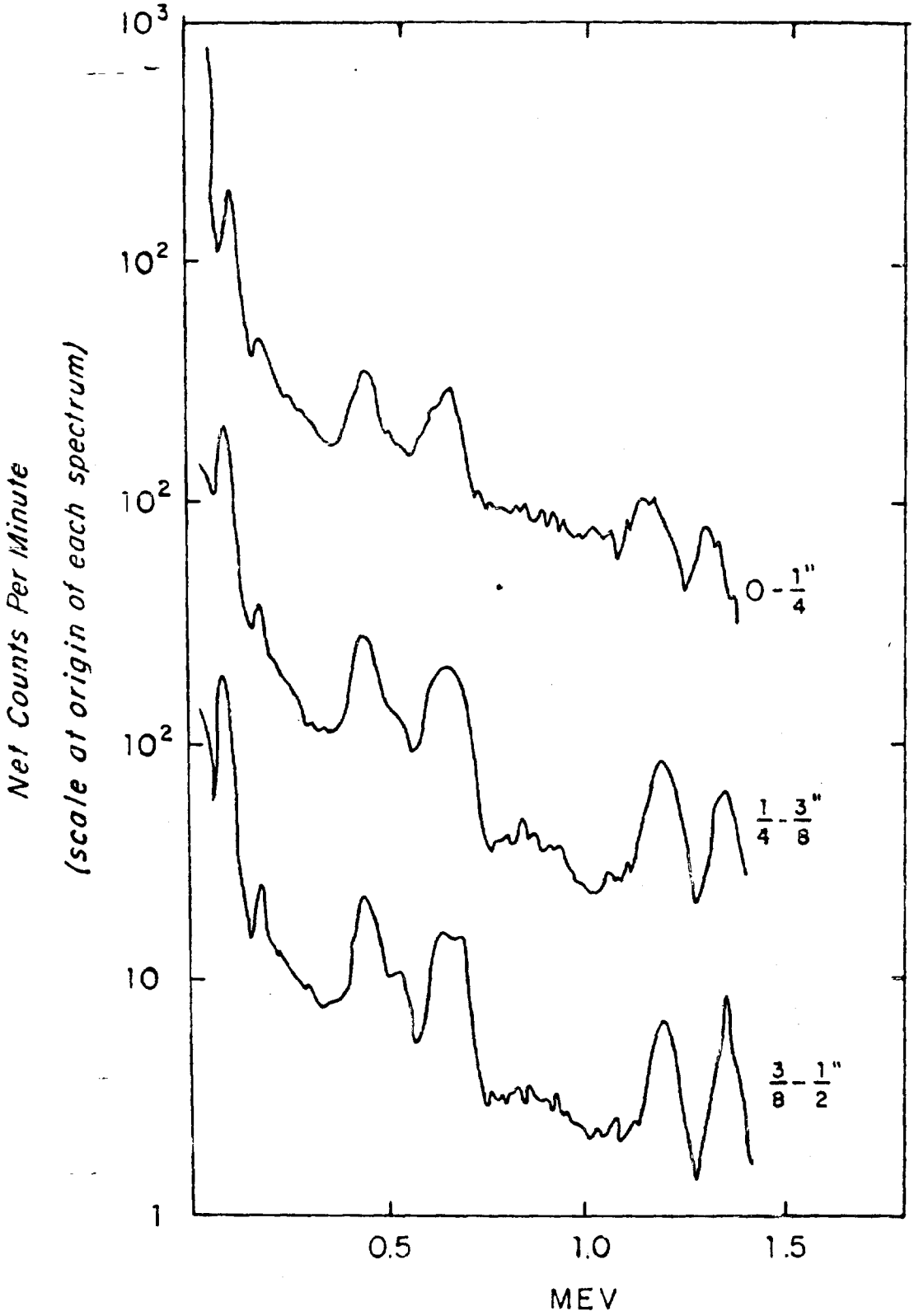


Fig. 7. Gamma-ray spectra of depth increments of old soil ta from the surface inch. The origin of each spectrum has been arbitrarily separated from the preceding one by a factor of ten to facilitate inspection of the figure.

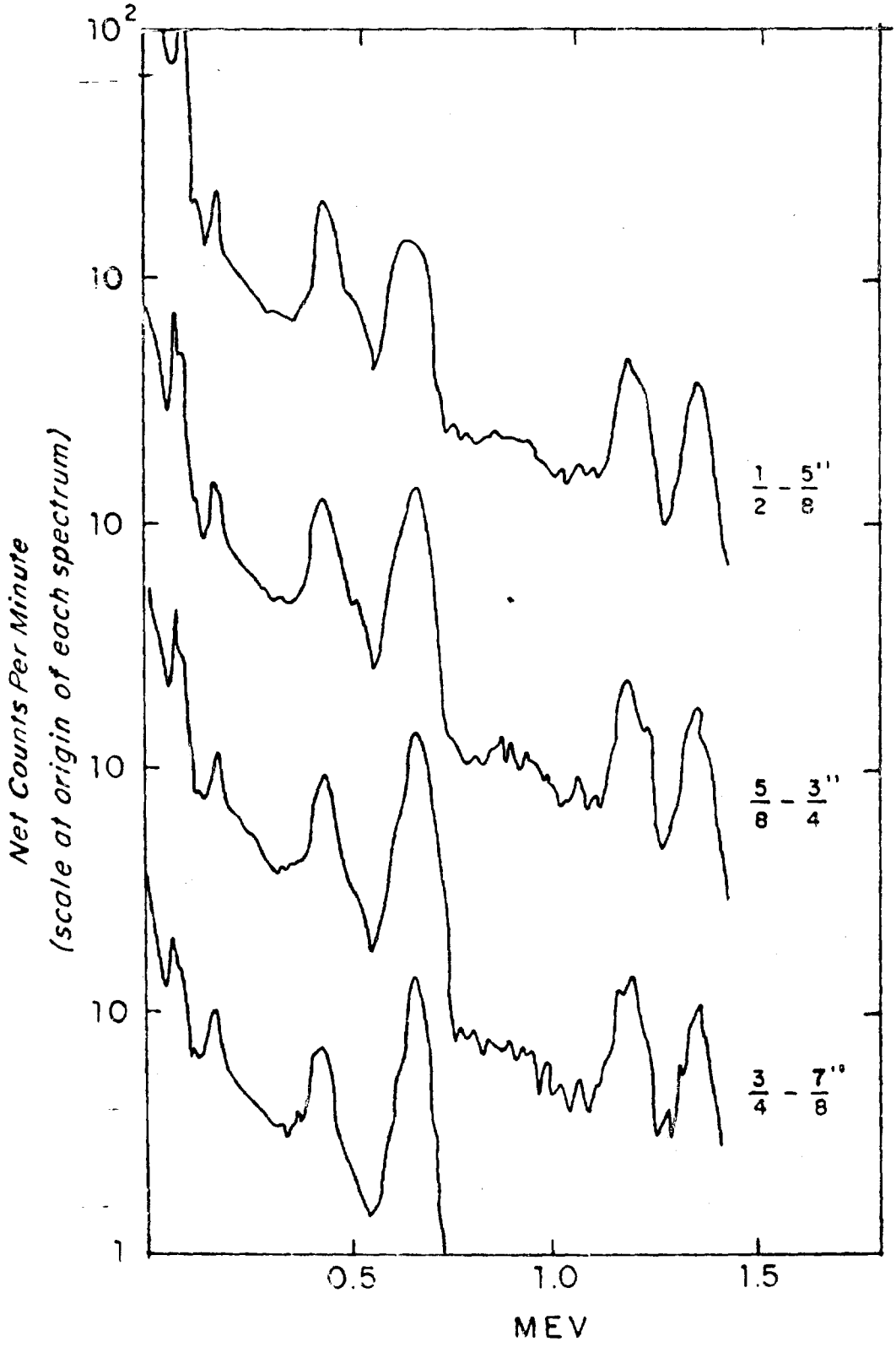


Fig. 7. (continued)

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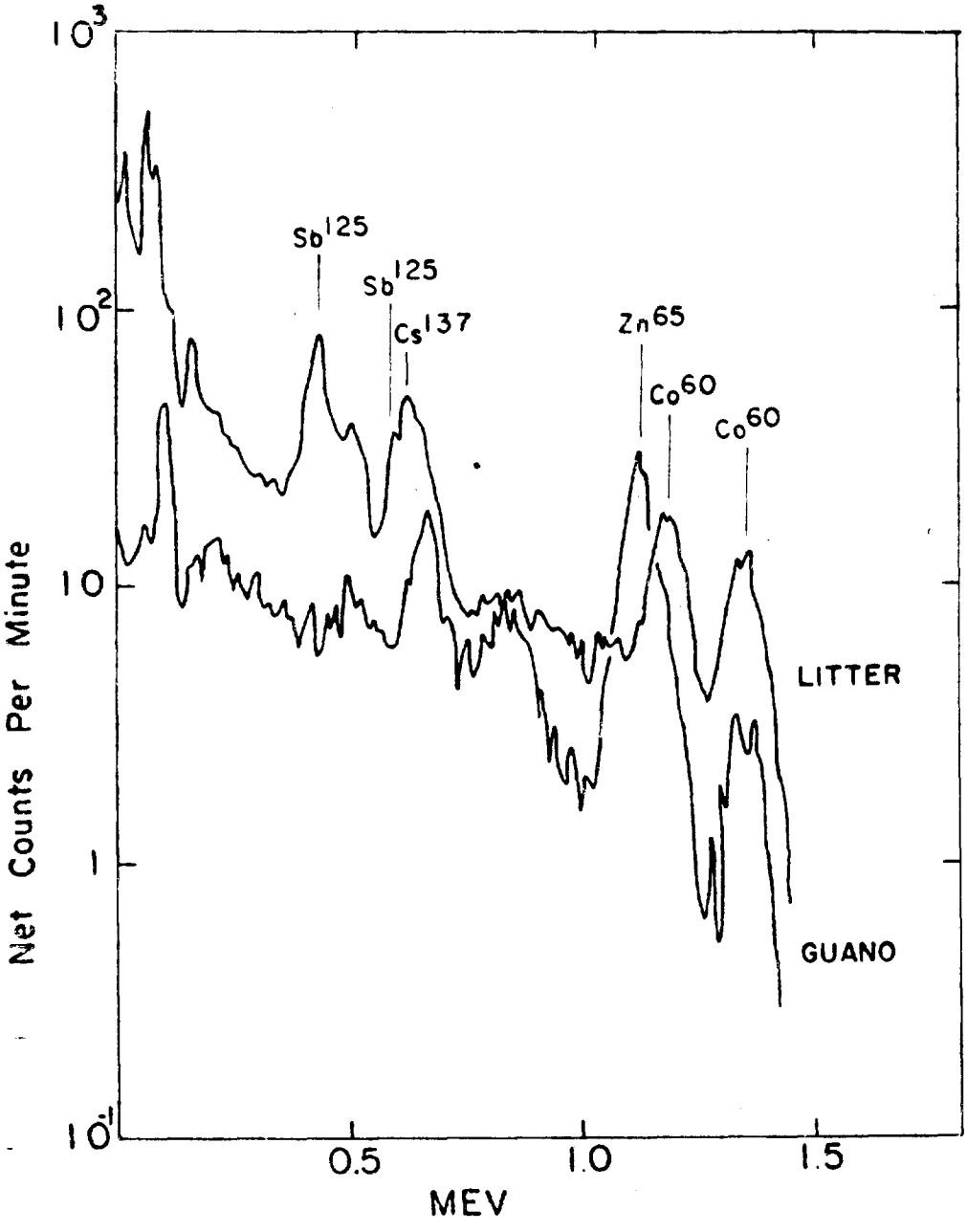

Fig. 8. Gamma-ray spectra of litter and guano collected in an area of old soil.

Zn^{65} peak. The presence of Zn^{65} is corroborated by the 0.5-mev peak. Fig. 8 also shows the gamma-ray spectrum of noddy tern guano collected in this area. The 1.12-mev peak predominates over the 1.17 peak of Co^{60} and the 0.51 peak of Zn^{65} is evident. The foliar contribution to the litter contains only Cs^{137} from among the gamma-emitters.

In undisturbed areas Cs^{137} and Sr^{90} are being deposited with the litter and are thus replacing at the surface some of the Cs^{137} and Sr^{90} lost by leaching. There is not sufficient data from the field work to determine whether there eventually will be a loss of these radionuclides from the soil-plant system, or a steady state (excluding physical decay of the radionuclides). Long-term experiments, under simulated field conditions, with monolith lysimeters and controlled and uniform addition of the radionuclides would define this point.

Young Soil

Fig. 9 gives the spectra of the 0 to 1-inch, 1 to 2-inch, and 9 to 10-inch increments of a young soil. Co^{60}, Zn^{65}, Ce^{144}-Pr^{144}, and Eu^{155} were detected only in the surface layers, and with increasing depth the 0.60 to 0.66-mev photopeak region of the spectra shifts toward the 0.60-mev peak of Sb^{125}. The spectrum of the 9 to 10-inch increment is compared with that of an Sb^{125} spike in Fig. 10, showing that the photopeaks of the soil and spike gamma spectra are identical.

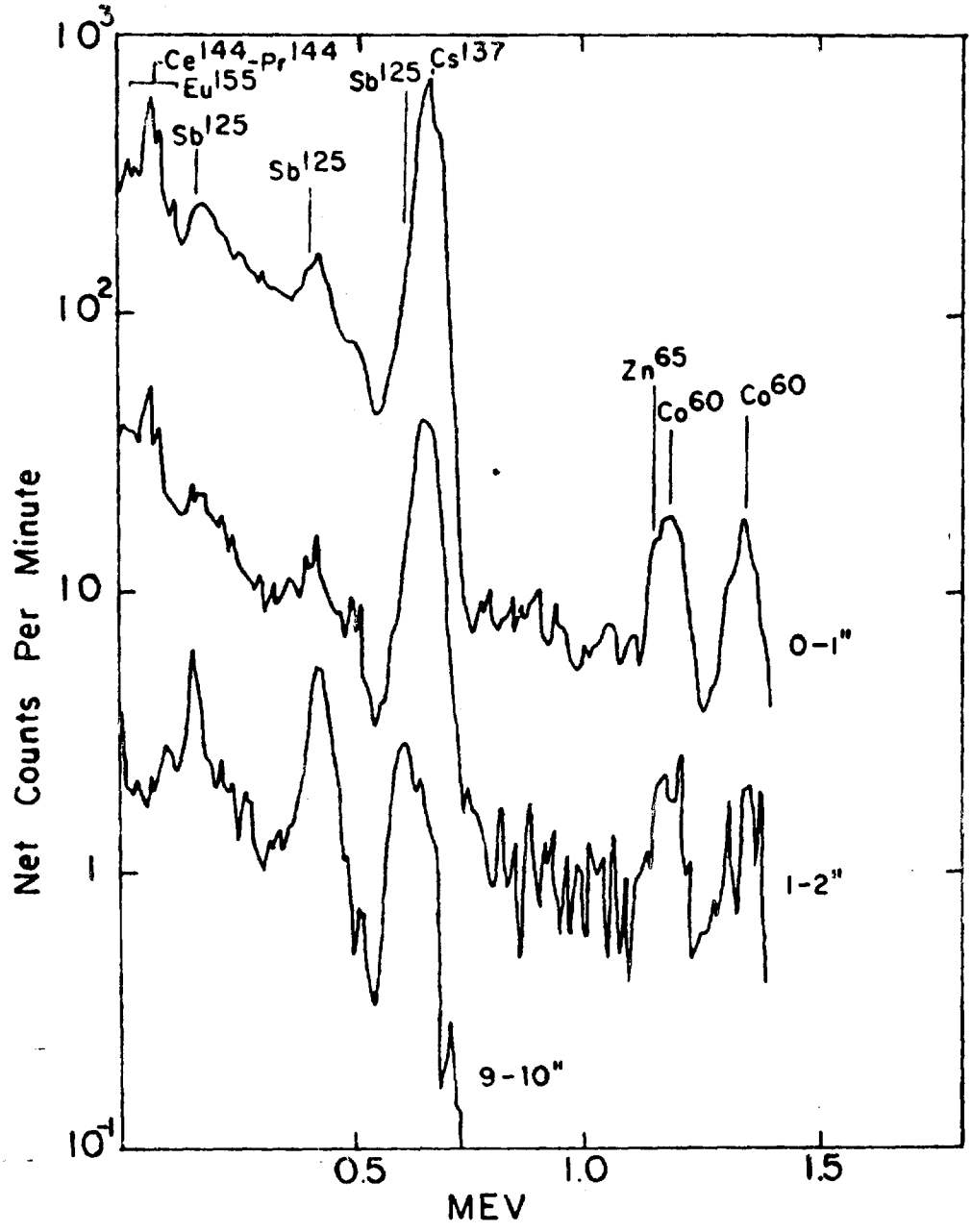

Fig. 9. Gamma-ray spectra of equal amounts of young soil from different depths.

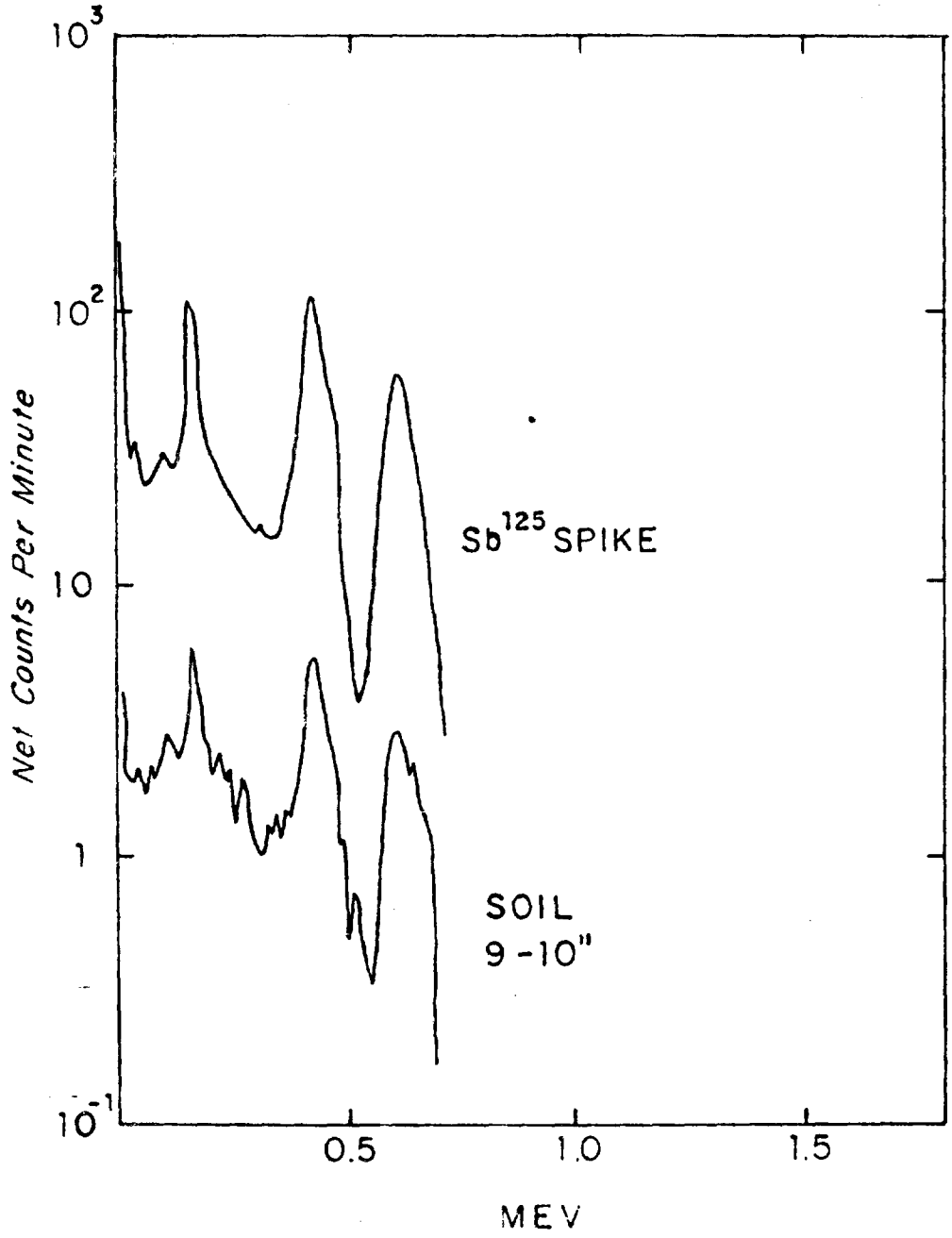

Fig. 10. Gamma-ray spectrum of young soil from a depth increment of 9 to 10 inches compared with the gamma spectrum of Sb^{125}.

The radionuclide content of leachates collected in the field agrees with these results (Cole et al. 1961). Leachates from young soil contained only Sb^{125} and Sr^{90}, while leachates from better developed soils contained mainly Sr^{90} and Cs^{137}, with traces of other gamma-emitters.

Depth Gradient of Sr^{90}

The relative Sr^{90} content of depth increments of the two soil types is given in Table IV. There is a rapid decrease in the amount of Sr^{90} with depth, and the differences in Sr^{90} content between soils probably are not significant. The extreme values from the results of Sr^{90} analyses of subsamples of replicate samples taken from small areas differ by a factor of more than ten (Table II). It is likely that the variability is due largely to the spotty nature of the distribution of the fallout radionuclides, which is evident in the radioautographs discussed below and from X-ray films that were exposed at the soil surface and just below the surface (Fig. 11), and to small differences in the characteristics of the soil within a single soil type.

Sampling by 1/8-inch increments in 1959 of an undisturbed old soil, (Fig. 4 and Table I) on Rongelap Island indicates a gradient of Sr^{90} levels in the top inch of soil (Table V). The levels in the second inch are about one tenth those in the

Table IV. Relative Sr^{90} content of depth increments collected in 1959 from two Rongelap Atoll soil types.

Depth in inches	Percent of total in surface ten inches	
	Old Soil (Pit 4)	Young Soil (Pit 6)
0 - 1	81.0	52.9
1 - 2	4.9	16.6
2 - 3	2.8	16.2
3 - 4	2.0	5.8
4 - 5	1.4	4.0
5 - 6	2.2	2.2
6 - 7	2.0	1.2
7 - 8	0.8	0.7
8 - 9	2.5	0.2
9 - 10	0.6	0.1
Total	100.2	99.9

Fig. 11. Radioautograph obtained by exposing no-screen X-ray film approximately one cm below the soil surface <u>in situ</u>.

Table V. Sr^{90} and Ca in the surface 1-1/2 inches of Rongelap soil, Rongelap Gravelly Sand, collected in September 1959.

Depth in inches	Sr^{90} Percent of total in top 1-1/2 inches	Ca mg/gm
0 - 1/8	27	292
1/8 - 1/4	26	291
1/4 - 3/8	16	315
3/8 - 1/2	10	337
3/4 - 7/8	3.2	346
7/8 - 1	2.7	348
1 - 1-1/8	2.0	306
1-1/8 - 1-1/4	2.6	338
1-1/4 - 1-5/8	1.5	329
1-5/8 - 1-3/4	2.2	328
1-3/4 - 1-7/8	3.6	351
1-7/8 - 1-1/2	2.2	369
Total	99.0	

first inch and remain at approximately the same level from 7/8 inch to 1-1/2 inches in depth.

RADIOAUTOGRAPHS

Young Soil

Undisturbed

Radioautographs of cores taken in 1963 show marked differences in the distribution of radioactivity. Fig. 12 shows photographs and the corresponding radioautographs of sections of cores of young soils from an undisturbed area (Fig. 2) and from an area subject to erosion (Fig. 13) and an old soil (Fig. 3). The radioactivity corresponds closely to the dark area in the photograph of the core from the undisturbed young soil. This dark area is composed almost entirely of a mixture of soil algae, forming a crust which has retained most of the fallout radionuclides.

Eroded

In an eroded area of young soil the radioactivity is associated with large coral fragments which are infiltrated with algae. This area is subject to erosion by both wind and water, which accounts for the coral fragments containing algae and radionuclides occurring below the soil surface. Radioactivity is not associated primarily with smaller particles as

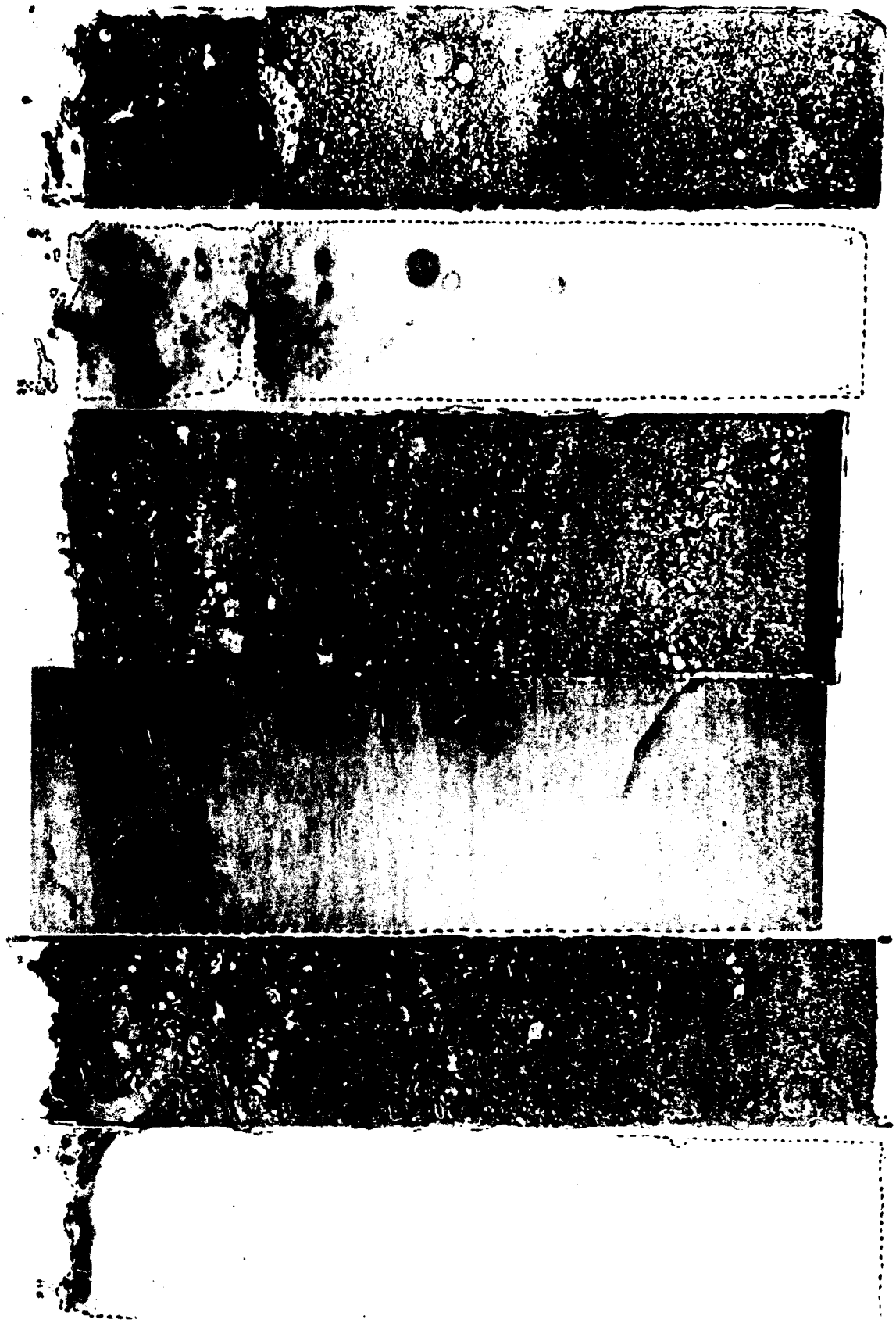

Fig. 12. Radioautographs and photographs of soil core sections. From left to right: undisturbed young soil, eroded young soil, old soil.

Fig. 13. An area of young soil subject to erosion.

in other soils, presumably because smaller particles have been eroded away since fallout occurred.

Old Soil

A radioautograph of a core from the old soil (Fig. 12) shows that the radioactivity extends to a greater depth and is more diffusely distributed than in the young soils. The surface litter contains little radioactivity compared with the soil surface.

Horizontal Movement

There is evidence of localized horizontal movement of radionuclides in the soil, at least near the surface, which in turn may affect vertical distribution. Fig. 14 is a photograph and radioautograph of a section of a soil core collected in old soil on Kabelle Islet. There is a darkened funnel-shaped area at the top center of the radioautograph and a corresponding darker area of high organic content in the core section. The amount of activity in the stem of the "funnel" appears to be too great to be accounted for by leaching from the surface directly above. Either a depression was filled with radioactive organic matter after fallout, or the depression was filled before fallout and the radionuclides were adsorbed when surface runoff water filtered through the funnel of organic matter. The latter interpretation seems more probable since gamma-ray spectra of

Fig. 14. Photograph and radioautograph of old soil showing correspondence of radioactive portions to organic matter.

layers of the funnel show only Cs^{137} and Sb^{125} in the deepest layer.

Buried Organic Layers

The organic matter in buried horizons may become important in the vertical distribution of radionuclides in the soils. No detectable gamma-emitters were found in buried horizons during the period 1958-1961. In September 1963 a core was taken through a buried horizon at 14 inches in old soil. Radioautographs of sections of the core show that a small amount of radioactivity has accumulated in the buried horizon. The diffuse distribution of the radioactivity in this case indicates that there is adsorption of the radionuclides from solution as water percolates through the soils, although the possibility of translocation by plants cannot be ruled out. Cs^{137} and naturally occurring K^{40} were the only radionuclides present in the buried horizon.

The accumulation of Cs^{137} in the buried horizons is of particular significance in terms of plant-soil relationships, because a proliferation of roots is often found in the buried horizons and Cs^{137} is the principal fallout radionuclide taken up by atoll plants.

Roots

Roots, which contain Cs^{137} and Sr^{90}, in most instances have higher levels of radioactivity than the surrounding soil except near the soil surface (Fig. 12). The influence of poplar roots on redistribution in a continental soil has been well demonstrated

by Witkamp and Frank (1963). It may be possible to distinguish between translocation within the roots and downward movement along root channels at Rongelap by comparing the ratios of radionuclides within the roots and in the immediately surrounding soil in core sections. For example, if there is greater movement along root channels, we would expect Sb^{125}, which is not absorbed by the plants, to be most abundant in the soil adjacent to the roots at depth, if there is appreciable channelization.

Pumice

Radionuclides are also adsorbed by pumice particles. No detailed morphological examinations of the soil sections have been made, but it is obvious in some core sections that a few of the larger "hot spots" several inches below the surface are associated with pumice fragments. The retention of radionuclides by pumice fragments may be of importance in considering soil-plant relationships in a few highly localized areas since proliferation of roots around pumice fragments has been observed (Sachet 1955; Kenady 1962). However, as pumice is rarely found beneath the surface, the effects of this material would not be generally important.

* * *

We can not explain the differences in distribution of radionuclides between soil types but assume that the greater retention

of all radionuclides, except Sb^{125} and Sr^{90}, at the surface of the young soil is associated with the algal crust. The observation that mosses and lichens collected from trees at Rongelap Atoll in 1961 show essentially the same gamma-ray spectra as the algal crust lends supporting evidence to this assumption. The retention of radionuclides by algal crust, mosses and lichens must be related to adsorptive surfaces or to the metabolism of the organisms, although it is impossible to determine from the field data the mechanism or combination of mechanisms involved. Similar observations have been made with arctic lichens (Palmer et al. 1964).

Since the algal crust at Rongelap Atoll has retained the radionuclides for nine years and from all indications will continue to do so for years to come, it is possible that it also retains a variety of mineral nutrients, thus providing a reservoir of nutrients in otherwise barren areas. This reservoir might be tapped upon invasion of the areas by higher plants and the concomitant activity of animals in mixing the upper layers of the soil.

Distribution of the radionuclides in the old soils arises from a combination of processes which are difficult to delineate. No doubt much of the movement is due to leaching and readsorption

of radionuclides moving through the soil. This is certainly true for Cs^{137} and Sr^{90}, which were abundant compared to other radionuclides in the leachates. It also was demonstrated by comparing the gamma-ray spectra of depth increments that there is a more rapid movement of Cs^{137} than of other gamma-emitters present. It should be recalled that the exchange capacity of these soils originates from the organic content, which, as is obvious from the photographs of the core sections and Table III, is far higher in the old than in the young soil. There is thus more opportunity for exchange and retention of radionuclides in old soil. Mechanical mixing, due mainly to the activity of land crabs, plays an important role in redistribution in the surface layers. This effect is obvious in areas where there has been active burrowing, and is probably occurring to a small extent throughout densely vegetated areas as is indicated by the presence of Ce^{144}-Pr^{144} and Eu^{155} in the litter. These radionuclides could only have come directly from the soil by upward mixing since they were not found in the vegetation which contributed significantly to the litter.

CONCLUSIONS

Different plant and soil environments on single islets have a different vertical distribution pattern of radionuclides from the same fallout material. The vertical distribution of

radionuclides in old soils is as follows, in order of greatest penetration: Cs^{137}, Sr^{90}, Sb^{125}, Co^{60}, Zn^{65}, Cr^{144}-Pr^{144}, Eu^{155} and probably other rare earths. In the young soils, consisting almost entirely of parent material, the positions of Cs^{137} and Sb^{125} are reversed and the other radionuclides appear to be more completely retained in the surface algal crust. The maximum concentration of fallout radionuclides remains at the soil surface, a few inches or less in depth, except in areas where there has been erosion. In the eroded areas large particles containing both soil algae and radionuclides are randomly distributed to a depth of a few inches.

There is some horizontal movement of radionuclides but such movement appears to be very localized and thus is of little consequence in the overall picture of distribution.

Cs^{137} and Sr^{90} are the principal radionuclides entering a cycle within the soil-plant system. Any loss from this system appears to be small (a fraction of one per cent per year), but a definite conclusion can not be drawn from the data.

SUMMARY

1. Rongelap Atoll received a single heavy dose of radioactive fallout in 1954.

2. The atoll soils are calcareous and contain no inorganic colloids; the exchange capacity is related to organic content.

3. Comparison of gamma-ray spectra of depth increments from old and new soils shows that Cs^{137} and Sb^{125} move most readily in the old soil; the principal gamma-emitting radionuclide moving in new soil is Sb^{125}.

4. Sr^{90} moves in both old and young soils.

5. The distribution of radionuclides at the surface is very spotty.

6. There is a vertical gradient in the distribution of radionuclides within the surface one-inch layer.

7. Litter redeposits Cs^{137} and Sr^{90} over the soil surface and bird droppings have added Zn^{65} and Co^{60}.

8. The principal reservoir of radionuclides in young soils is the surface algal crust.

9. In eroded areas radioactivity is associated with large coral fragments, which are infiltrated with algae.

10. There is some localized horizontal movement of radionuclides in old soils.

11. Buried organic horizons contain more Cs^{137} than adjacent soil layers.

12. Roots are generally more radioactive than the surrounding soil except at the soil surface.

13. Pumice particles in the soil adsorb radionuclides.

14. Mechanical mixing by animals in old soils and by erosion in young soils results in a redistribution of radionuclides in the surface layers.

15. Cs^{137} and Sr^{90} are the principal radionuclides entering a cycle within the soil-plant system.

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MINERAL CYCLING IN A DOUGLAS FIR FOREST STAND*

HANS RIEKERK and STANLEY P. GESSEL
College of Forestry, University of Washington, Seattle, Washington

(*Presented by* H. RIEKERK)

Abstract—This study of the movement of mineral elements through the forest soil confirmed several observations made by other investigators. The early fall rains cause a flushing of decay products accumulated during the summer. Movement of the various mineral elements depends very much on their chemical characteristics, and on the fixation properties of the soil system.

^{32}P release from the forest floor ranged from 2 to 14 per cent, ^{86}Rb release was 0.01–1.88 per cent and ^{45}Ca release was 0.06–0.86 per cent for similar periods of leaching. ^{45}Ca release over periods of leaching two to three times longer amounted to 0.1–4.1 per cent. Release of ^{32}P from the surface soil was about 2800 times less than that from the forest floor. ^{86}Rb was 2200 times less and ^{45}Ca was about 160 times less than the release from the forest floor. Release patterns of ^{45}Ca indicated a slow movement as compared to ^{32}P and ^{86}Rb.

Uptake of ^{32}P by the trees was evident within a week after application. ^{32}P uptake was about ten times more during spring than during fall. Uptake of ^{86}Rb was evident after about a month during the spring season, but was not observed during fall. Uptake of ^{45}Ca was not significant.

INTRODUCTION

RESEARCH in forest productivity and fertilization has indicated the importance of nutrient dynamics.[1,2] The entry of radioactive fallout products into the cycling processes of biological importance has received considerable attention.

COLE[3] reported on a study of mineral cycling in western Washington. He found marked differences in the mineral retention of the major nutrient elements by the forest floor. The annual release of potassium was 56 per cent of the total capital present in the forest floor. Calcium release was 7 per cent, nitrogen 2 per cent and phosphorus 1 per cent of the total. Comparison of these releases to the input by litterfall indicated an accumulation process. Leaching prior to usual collection of litterfall was deduced to be 80 per cent of the potassium and 40 per cent of the magnesium involved with the transfer. Maximum rates of release were reported for fall. Abrupt changes in the ecosystem by clearcutting and fertilization modified these general release patterns.[4] Except for calcium these treatments had no effects on the movement of mineral elements deeper in the soil. Nearly all the elements were retained within the rooting zone.

Studies conducted at the Oak Ridge National Laboratory, Tennessee, with radio-cesium tracers in White Oak and Tulip Poplar indicated a high mobility. In particular, freshly fallen litter released relatively large amounts of cesium in a short time.[5,6]

It is the purpose of the present study to investigate in more detail the cycling processes as reported by COLE and GESSEL. The use of radioactive tracers makes it possible to measure velocities, and more important, to distinguish newly arrived nutrient elements from the amounts already present in the various components of the ecosystem. For example, the calcium content of the soil solution is a mixture of calcium released by the soil and calcium passing through directly from the forest floor.

For this study three contrasting mineral elements in carrier solutions were tagged and applied to the forest floor or surface soil.

* This study is based on work performed partly with support from a National Science Foundation grant to S. P. GESSEL, and partly with support from the Radiation Biology Laboratory (AEC Contract AT(45-1)1385), University of Washington, Seattle, Washington.

Phosphorus was selected in addition to the monovalent potassium and divalent calcium cations. These three elements can easily be tagged with ^{32}P, ^{86}Rb and ^{45}Ca for tracer studies.

This paper will report only the radiological aspects of movement of the tracers through the forest floor and surface soil, and up into the trees.

STUDY AREA

The study is conducted in close cooperation with the Fern Lake Program of the Radiation Biology Laboratory, University of Washington, Seattle, Washington.[7] Fern Lake is located about 50 miles southwest of Seattle, Washington. The climate is moderate with a mean annual temperature of 50°F and an average annual precipitation of 45 in. The study plots are located close to the lake in a dense stand of 40-y old Douglas fir trees (*Pseudotsuga menziesii* (Mirb.) Franco) about 50 ft high. The fairly level soil is an Alderwood-like gravelly sandy loam originated on glacial till. The forest floor is about 1 in. thick and has a 50 per cent moss cover. The A_1 horizon is 4–5 in. deep. The pH throughout the profile is about 5.3, and the cation exchange capacity decreases from 12 meq/100 g at the surface to 5 meq/100 g at a depth of 2 ft.[3]

METHODS

Each plot consists of a central dominant tree without close competition from other dominants (See Fig. 1). The forest floor and/or surface soil of 225 in.^{2} areas arranged around the central tree were sprayed with tracer solutions of known specific activities. Tension lysimeters, installed under the forest floor and 6–8 in. in the surface soil, collected soil solutions. The tension lysimeter consists of a porous plate that can be readily inserted within a forest soil with a minimum of disturbance to the overlying system. A tension placed against the porous plate by a controlled vacuum system eliminates the soil solution and air interface problem inherent in the drainage of most lysimeter designs.[8]

Uptake patterns of the tracers by the central tree were obtained by periodically sampling young foliage of midcrown positions. A steel tower erected alongside the tree facilitated access to the crown. The return movement of mineral elements was sampled with litterfall troughs placed radially from the tree, and a rubber channel sealed around the stem. Rainwash solutions from the troughs and stem were led through mixed bed ion exchange resin columns which contained porous plastic filters to prevent clogging of the resins. Constant siphontube flowmeters were used to monitor the flow of solutions at several sampling points. The discharge of the flowmeter by siphon action cools a positive temperature coefficient resistor which then causes a relay to activate an event recorder. Incoming precipitation was measured by rain gauges placed on two of the towers. Forest floor and surface soil temperatures were recorded continuously at one location in the forest stand. The radioactivity of a few lysimeter solutions was recorded continuously with the aid of throughflow GM-tubes. A control plot to monitor background activity due to radioactive fallout products in the samples was maintained throughout the study.

During 1964, 12.5 per cent of the total radioactivity per plot was applied on the forest floor and 87.5 per cent directly on the surface of the mineral soil. The forest floor was separated and lifted off the application area with a board and replaced after treatment of the exposed soil surface.

The solutions, exchange resins and tree crowns were sampled periodically. The solution volumes were determined by weighing to prevent cross contaminations. Excess radioactive leachates were evaporated in the field in a barrel with a floating heating element.[9] A mercury switch disconnected the heating element when the solution dropped below a given level.

One-liter samples were evaporated and oxidized in the laboratory and assayed for radioactivity. The exchange resins were eluted with 2–3 volumes of 10 per cent HNO_3. The eluates were evaporated to dryness, oxidized and assayed for radioactivity. The foliage samples were slowly ashed at 450°C, taken up in N HNO_3, filtered and assayed. Calcium in the foliage samples of 1964 was precipitated out by the oxalate method[10] to reduce the radioactivity due to fallout products.

All samples of 1963 were assayed for beta activity by the Radiation Biology Laboratory. During 1964 the gamma radiation of ^{86}Rb was measured using a 4-in. scintillation crystal and a multichannel analyzer. Data on recovery

Table 1. Rates of application of ^{32}P, ^{86}Rb, *and* ^{45}Ca

	1963				1964			
	Spring		Fall		Spring		Fall	
	mCi/in.²	Total mCi	mCi/in.²	Total mCi	mCi/in.²	Total mCi	mCi/in.²	Total mCi
	0.018	12	0.016	17	0.056	102	0.056	100
	0.017	10	0.023	25	0.111	200	0.111	202
	0.009	6	0.010	11	0.018	32	0.017	30

ed elements is given in the tables and figures as parts per million (ppm) and expresses the fraction of the radionuclide recovered in a year. This can be converted to per cent by dividing by 10,000.

RESULTS AND DISCUSSION

Table 1 presents information concerning the rates of application of the tracers. Larger amounts have been applied during 1964 because of the emphasis on the measurement of uptake by trees and subsequent return to the soil with throughfall and rainwash.

The count data have been corrected for decay and background radiation due to fallout products. The corrected data were converted to counts per min/ml, and to ppm of the remaining radioactivity after subtraction of leaching losses. Table 2 presents the totals leached during the various periods of sampling.

The ratios between the amounts released by the forest floor and the surface soil during similar periods of leaching indicate the degree of retention in the surface soil. Phosphorus and rubidium appear to be retained more than calcium. For phosphorus this might be related to the high iron content of the soil as indicated by the presence of iron concretions throughout the surface soil. A potassium deficiency of trees in the general area may indicate a high potassium fixation in the soil,[11] and very likely also rubidium. Calcium has also been reported to move through the soil at a low rate.[3]

Some variation between the levels of total

Table 2. Parts per million of* ^{32}P, ^{86}Rb *and* ^{45}Ca *released by the forest floor and surface soil*

	1963					1964			
	Spring		Fall		Average	Spring		Fall	
Date of application	ppm	days	ppm	days	ratio†	ppm	days	ppm	days
^{32}P Forest floor	149,000	4	23,300	48		37,200	78	38,200	133
Surface soil	31	4	26	72		0	78	4721	133
Ratio	4800		900		2800	—		8	
^{86}Rb Forest floor	18,800	4	13,400	135		—	—	99	133
Surface soil	5	4	19	98		—	—	8	133
Surface soil	—	—	—	—		—	—	1	133
Ratio	3750	—	710	—	2200	—		12–99	
^{45}Ca Forest floor	572	4	662	72		848	78	8650	132
Forest floor	41,200	454	1020	251		16,400	290	—	
Surface soil	2	4	22	72		0	78	27	132
Surface soil	2	454	49	251		2080	320	—	
Surface soil	—		—			0		11	132
Surface soil						731	320	—	
Ratio	286		30		160	—		320–790	
Ratio	20,500		82			8–22		—	

* Parts per million $\times 10^{-4}$ = per cent.

† Partial application to the mineral soil makes direct comparisons invalid for 1964 data.

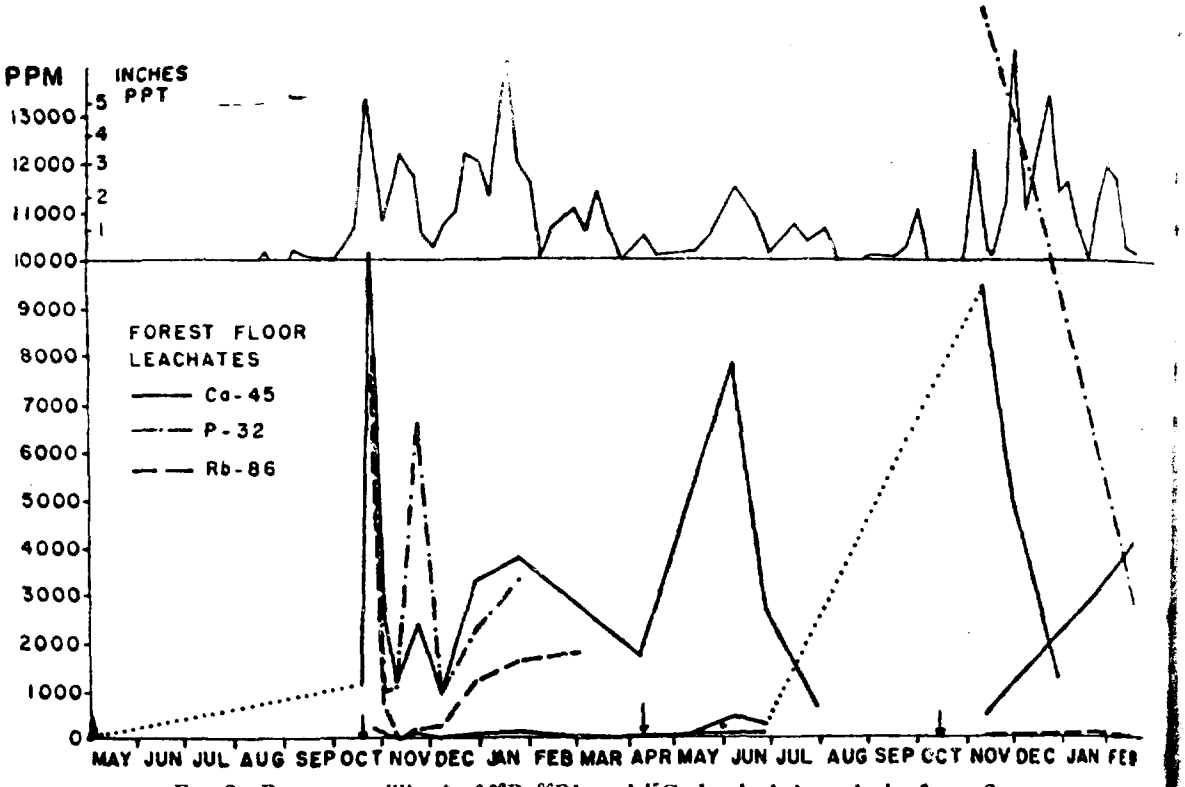


FIG. 2. Parts per million* of ^{32}P, ^{86}Rb and ^{45}Ca leached through the forest floor.

release might be explained by the total amount of forest floor and surface soil above the lysimeters. This effect is particularly noticeable in the release patterns of calcium applied during the fall of 1963, and rubidium applied during the fall of 1964 over the same lysimeter (see Fig. 2).

The data as presented in Figs. 2 and 3 indicate a pronounced flushing effect immediately after the first fall rains. This is particularly so in a few of the calcium collections made just before and after these rainstorms. However, a state of nonequilibrium of the newly applied tracers may have confounded the picture. This is indicated by the very high values of ^{32}P and ^{86}Rb collected under the forest floor for only a few days after application during the spring of 1963 (see Table 2).

The release of ^{32}P from the forest floor, as presented in Fig. 2, shows a decreasing trend in contrast to ^{86}Rb. This might be explained by a reduced microbiological activity due to the lower temperatures of the season. However, the effects of continued phosphorus fixation might be a significant factor. It is also known that potassium is more mobile when the soil system becomes sufficiently moist.[12]

The release patterns of calcium from the forest floor show considerable fluctuations, but with a generally increasing trend. The absence of a fall-flushing effect of the newly added ^{45}Ca may indicate a slower movement through the forest floor. This is more clearly indicated by the release patterns of calcium newly added to the surface soil (see Fig. 3).

Figures 4 and 5 present the data expressed as counts/min/ml leachate. Here, the dilution effects of incoming precipitation may be observed. For example, the ^{45}Ca concentration of the forest floor leachates sampled just before and after the first rains of fall 1963, and of the surface soil during fall 1964, is decreasing (Fig. 5); but the total amounts (Fig. 2) show a sharp increase. A similar trend may be observed for the release

* Parts per million $\times 10^{-4}$ = per cent.

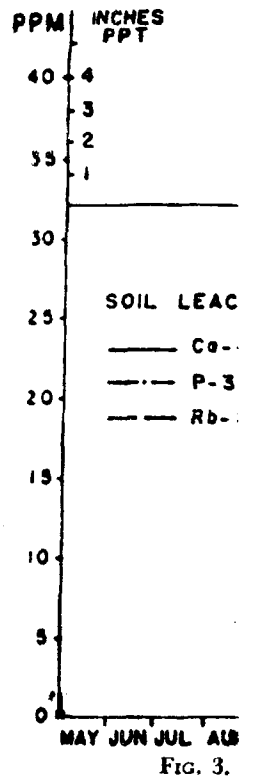
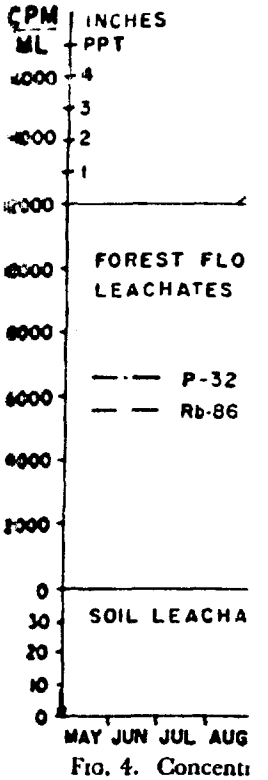


FIG. 3.



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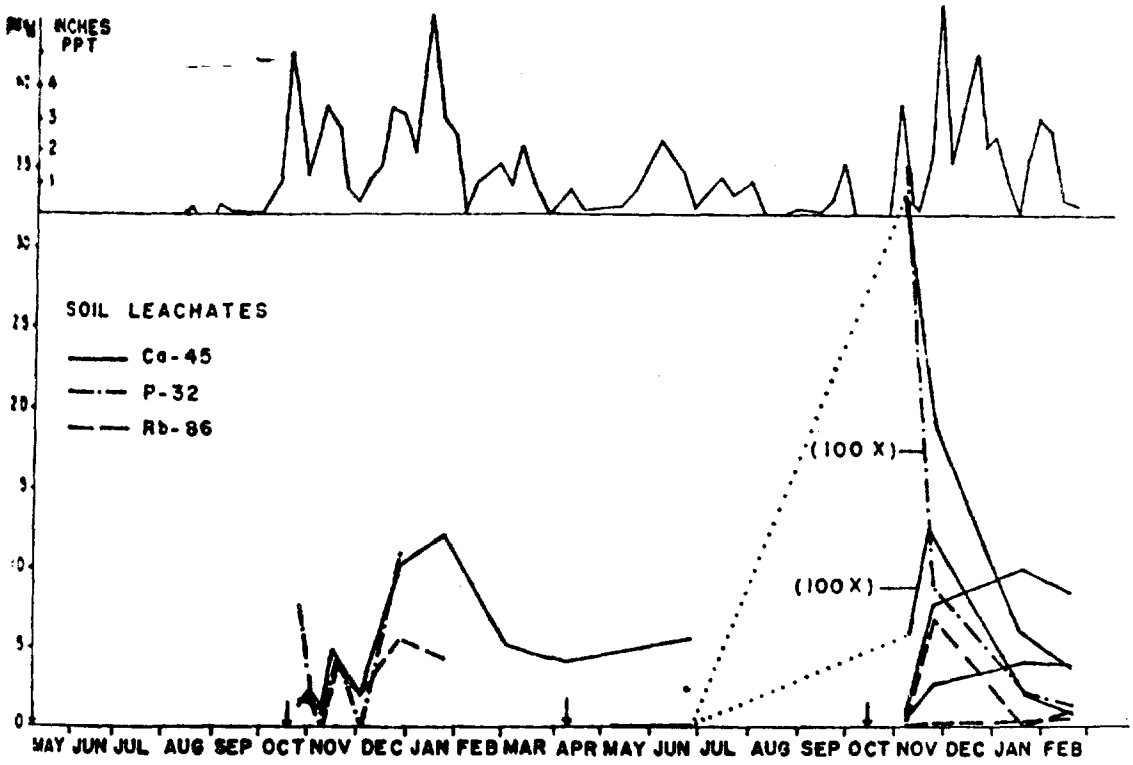

FIG. 3. Parts per million of ^{32}P, ^{86}Rb and ^{45}Ca leached through the surface soil.

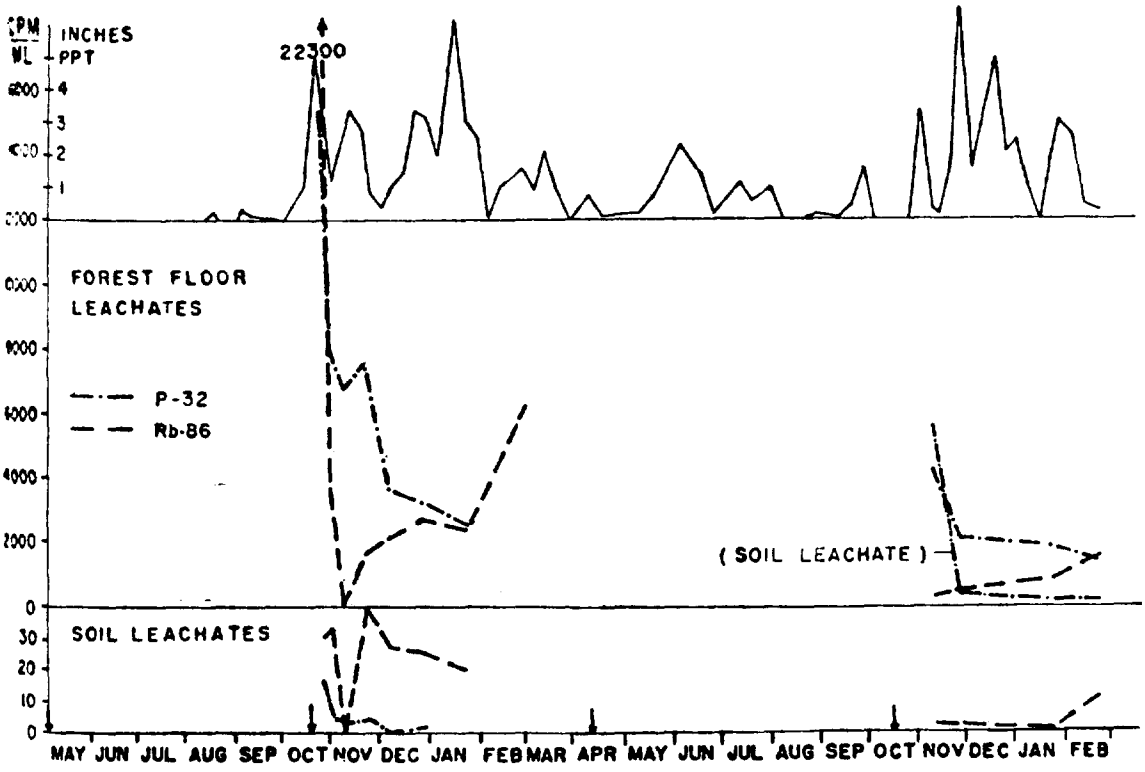

FIG. 4. Concentration of radioactivity of ^{32}P and ^{86}Rb in forest floor and surface soil leachates.

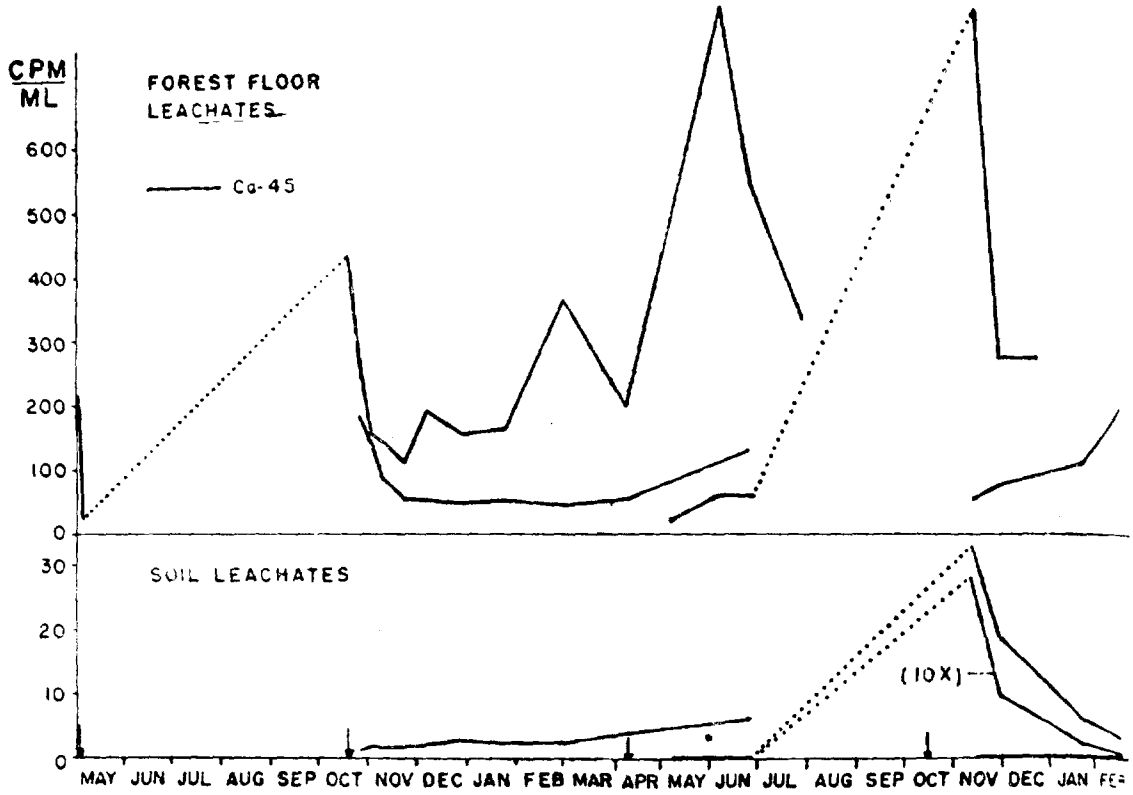

FIG. 5. Concentration of radioactivity of ^{45}Ca in forest floor and surface soil leachates.

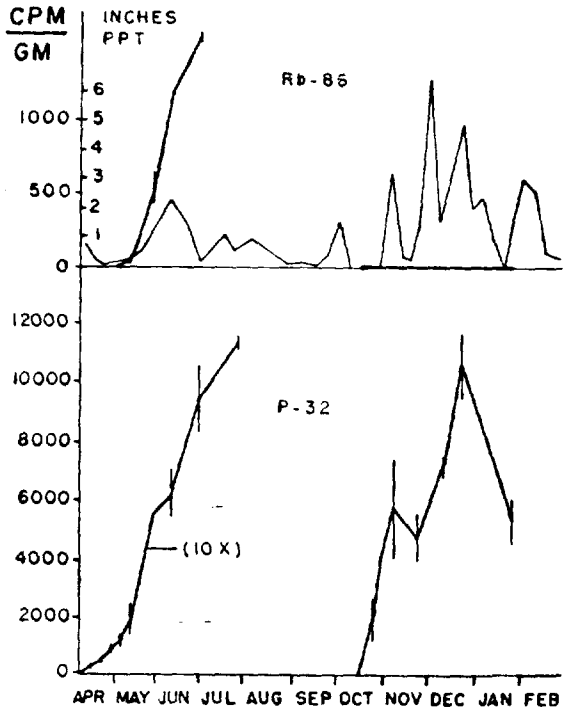

FIG. 6. Uptake patterns by Douglas fir trees of ^{86}Rb and ^{32}P.

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tern of ^{86}Rb from the surface soil during the l. of 1963 (Fig. 4).

The uptake data are presented in Fig. 6. The ^{86}Rb appeared in the trees within a week after application. The uptake during the spring season was about ten times that for the fall season. The uptake of ^{86}Rb was much slower and was significant about a month after application to the soil. The fall application of ^{86}Rb did not appear in the foliage samples. The limiting factor is most likely to be located in the soil system, because the phosphorus uptake indicated a sufficient biological activity of the trees at that time. The count data for ^{45}Ca after chemical separation did not show a significant activity.

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