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PROMPT TRANSFERS OF CESIUM-137 TO THE SOILS OF A TAGGED *LIRIODENDRON* FOREST[1]

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Abstract. An inventory of total Cs^{137} distribution in a 500-m^2 tagged tulip poplar (*Liriodendron tulipifera*) forest at the Oak Ridge National Laboratory showed transfer from trees to soil during the first growing season. About one-third of the maximum amount of radiocesium found in the tree canopy during June was transferred to the forest floor and soil; the remainder moved back into woody tissue (stem and roots) before leaf fall. Losses of radiocesium from the foliage and stems into the plot area through litterfall (55 $\mu c/m^2$) and rain leaching (13 $\mu c/m^2$) accounted for almost 8% (68 $\mu c/m^2$) of total Cs^{137} (934 $\mu c/m^2$) introduced into tree boles. However, soil (including roots) and litter sampling at the end of the growing season indicated a much larger quantity (414 $\mu c/m^2$ or 45% of initial input) already in the surface 30-cm layer of mineral soil. Of the total Cs^{137} activity found within the mineral soil layer, about half can be physically associated with the tagged trees through removal of roots from soil samples and cumulative measurement of surface income at time of soil collection. Much of the remainder can be accounted for by contact exchange, root exudation, root death, and leaching of roots by soil solution.

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Introduction

Many workers on mineral nutrient circulation within a forest have been concerned with the build-up and decay of organic matter on the surface of the forest floor. This material in turn furnishes surface income of material to the underlying soil. Less attention has been given to the underground transfer of mineral elements from organisms to soil, even though turnover of material from roots may be quite important as compared with the aboveground transfer.

One reason for greater study of the aboveground transfer from woody plants to soil has been the convenience in sampling components that are readily seen. The recent availability of radionuclides has made it more feasible to make quantitative estimates of transfer rates of elements from plants to soils, even those transfers which are associated with roots. This paper concerns one objective of a forest tracer experiment at Oak Ridge, Tennessee: to give a quantitative comparison between the surface and subsurface transfers of Cs^{137} to soil from inoculated trees in an important kind of forest community dominated by tulip tree or yellow poplar (*Liriodendron tulipifera*).

Results confirm the importance of transfers from tree roots and contribute to a broader objective of this experiment: to interpret the transfer of a long-lived radionuclide from dominant trees through all parts of an ecosystem. Related investigations help to identify and measure processes which connect the several compartments of such a system. An introductory report (Auerbach, Olson, and Waller 1964) and a discussion of equations for Cs^{137} transfer during the first summer (Olson 1965) have emphasized the changes in foliage concentrations due to a shifting balance between income and loss in the forest canopy. The present report follows the resulting changes in the forest floor and the soil through the first autumn (1962) and explains the reasoning behind our budgets of income and loss. Brown (1964) has discussed the organic binding of radiocesium inside the tree tissues at the end of the first year. Witkamp and Frank (1964) followed the first-year sequence of release of Cs^{137} and temporary immobilization and transfer by fungi and bacteria. Reichle and Crossley (1965) have reported preliminary results on the animal food chains which derive Cs^{137} from the forest floor compartments discussed here, and which return Cs^{137} to the litter and soil in excrements. Studies of radiation dosimetry in the forest and of the transfer of Cs^{137} to a new generation of seedlings via the seeds of tagged trees have been carried on by Witherspoon and Brown (1965).

Methods

A 20- by 25-m area of a mesophytic forest on colluvial soil (Emory loam) in a flat-bottomed sinkhole depression was tagged with $Cs^{137}Cl_2$ (carrier free); this substance was introduced into the boles of 35 trees during late May 1962 by methods described elsewhere (Auerbach, Olson, and Waller 1964). Three techniques for soil sampling were used because: (1) a large number of small core samples established a meaningful inventory in spite of wide variation between individual samples; (2) larger volume samples estimated more accurately the distribution of Cs^{137} between the different soil and root components; and (3) a sample system of boxes with soil slabs helped to interpret surface vs. subsurface sources of income to the soil.

Extensive core sampling

Exploratory sampling in July 1962 indicated that a large variation in Cs^{137} concentration existed between individual samples (Olson 1965, Table 3). Because of the uneven distribution of Cs^{137} within the soil, 10 randomly located samples were used in each of twenty 5- by 5-m subplots for an average inventory of this radionuclide in October. Use of a 2-cm diameter soil tube sampler permitted sampling to 30-cm depth with little destruction of the forest floor and provided a sample size that could be counted in an automatic changer-scaler (Packard Auto Gamma Counter-210). Each of 200 cores was segmented into six 5-cm layers giving a total of 1,200 end-of-season samples.

Because of the small cross section of the tube, the litter layer (undecomposed leaf material) was excluded in this sample system, although some partially decomposed organic debris may be included along with the 0- to 5-cm soil layer.

Components of decimeter slabs of soil

To determine the concentration of activity within the soil components (rock, roots, and soil), 20 decimeter-square profiles were taken to a depth of 10 cm into mineral soil, and each profile was subdivided into five units (litter layer plus four 2.5-cm soil layers). The soil layers were hand-separated into three components ($>$ 1 mm roots, $>$ 2 mm rock, $<$ 2 mm soil component) using nested sieves and forceps. Soil components were separated further by using the difference in specific gravity between mineral soil and fine organic matter.

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system by root exudation, die-off of small roots and root hairs, transfer to mycorhizal fungi, and leaching of roots by the soil solution. All of these processes are known to occur, but their quantitative or relative importance in transfer of materials to mineral soil is not very well documented.

For this reason the early tagging results are of special interest for evaluating the relative importance of belowground and aboveground transfer of mineral elements. If that portion of Cs^{137} remaining in the mineral soil residue after the nitric acid treatment (Fig. 7, line 6) is considered to be the only fraction completely disassociated from roots at the time of sampling, then the subsurface transfer of radiocesium out of the roots is about as great as the aboveground transfer. However, later nitric acid leaching experiments with Cs^{137}-spiked soils suggest that a major portion of the Cs^{137} associated with the nitric acid leachate (Fig. 7, lines 4, 5) was also associated with mineral soil colloids. This would indicate that the turnover of Cs^{137} (and probably other alkali metals) from tagged yellow poplar trees is greater belowground than aboveground.

Although the promptness of underground turnover just described appears surprising at first, the possible importance of such transfer has been suggested by several workers and possible explanations for transfer have been offered. Stenlid (1958) and Fujiwara and Iida (1956) discuss possible losses of materials from roots due to excretion and leaching. Based on estimates of Orlov (1955), Remezov (1958, 1959) emphasized the possible mineral and organic turnover due to production and death of roots throughout the season. He estimated that .4 metric tons per hectare, half of the amount produced, died per year in a 50-year-old oak stand. He also indicated that the death of small roots < 0.3 mm in diameter with high mineral content affects the mineral cycle significantly. Although they did not attempt a quantitative budget, Woods and Brock (1964) inferred that root transfer of minerals may be greater than is normally thought, and they cited further papers on the role of root exudates and mycorrhizal fungi as avenues of transfer to the rhizosphere and other species.

Discussion and Conclusions

An inventory of total Cs^{137} distribution among the tree and soil compartments of a tulip poplar forest (tagged May 1962) was constructed from data collected during the first growing season. A measurable influx of the radiocesium to the ground was recorded only 3 days after inoculation and over 40% of the initial inoculant was found in the litter, soil, and smaller root components by October (Fig. 1). The maximum burden found in the canopy about 4 weeks after inoculation (248 $\mu c/m^2$ of ground area) was reduced to 117 $\mu c/m^2$ by late September. Of the 131 $\mu c/m^2$ reduction, export of Cs^{137} from foliage to woody tissue by mature and senescent leaves accounts for almost 75%, rain and dew leaching about 7%, and early drop of leaves 18%. October losses were about 68, 3.6, and 49 $\mu c/m^2$ by translocation, rainout, and leaf drop respectively.

The important paths of transfer of cesium-137 from tagged trees to the forest floor included not only rain leaching and leaf fall, but especially passage through the roots. Transfer to the untagged understory vegetation by foliar interception of radiocesium leached from overhanging crowns of tagged trees is initially far more important than uptake from Cs^{137} in the soil (Waller, Olson, and Cristofolini 1965).

This study, like a previous one using small white oak (*Quercus alba*) trees (Witherspoon 1964), thus provides quantitative measures confirming the rapid translocation of Cs^{137} throughout major plant components of a forest. By contrast, cesium is a relatively immobile element when in direct association with the soil and tends to accumulate there (cf. Nishita, Steen, and Larson 1958, Titlyanova and Timofeyeva 1959, Schultz, Overstreet, and Barshad 1960, Lai, Nuang, and Chang 1961).

Woods and Brock (1964) concluded that the rate of transfer from roots to soil was much faster for P^{32} than for Ca^{45}. Wittwer, Bukovac, and Tukey (1963) listed comparative basipetal movement within plants from nutrients applied on foliage in four categories: (1) highly mobile elements —nitrogen and alkali metals; (2) mobile elements—phosphorus, chlorine, and sulphur; (3) partially mobile—zinc, copper, manganese, iron, and molybdenum; and (4) immobile—boron and alkaline earths.

If this relationship between basipetal mobility within plants is similar to that of transfer *from* root systems, then one would expect the other members of the alkali metals to be transferred at high rates relative to the members of the alkaline earth group such as calcium and magnesium. Alkaline earth elements should be among the least mobile elements in their contribution to soil by downward movement through plants, but they should have a relatively greater contribution to the cycle through surface income including leaching and leaf fall of the aerial components.

If such regularities in transfer rates apply widely

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In a flotation process, 50-g samples of soil were placed in a 250-ml erlenmeyer flask with about 150 ml of distilled water. Compressed air bubbled through the soil-water solution freed the larger organic material from the soil allowing it to float to the surface. The organic material (< 1 mm roots) was then removed and the concentration was determined and corrected for Cs^{137} activity measured in water.

Chemical leaches on the remainder of the soil showed differences in ionic bonding or physical entrapment of Cs^{137} in the living and dead organic material of the above soil component. Three reagents were applied: tap water; 1 N HCl (Jackson 1958, sections 5–10 to 5–12); and 1 N nitric acid (sections 6–83 and 6–84). Leachates and residues (including filter paper) were radioassayed for Cs^{137} with a gamma spectrometer.

Forest floor sample boxes

Surface litter and soil-sample units (contained in plastic sandwich boxes as described by Olson and Crossley 1963) provided one measure of surface vs. subsurface income through exclusion of roots. Interpretation of the distribution of surface income would otherwise have been very difficult since so much Cs^{137} reached undisturbed soil through roots. Also, independent estimates of surface income from rain leaching and from litterfall discussed below provided a basis for interpreting income. Repeated measurement of boxes and of surface income helped to convert a static inventory into a running budget of accumulated transfers for this forest throughout the first growing season.

Each plastic box had four round holes cut in the bottom to provide access and/or escape of small soil insects and moisture. Plastic screen wire with about 1-mm openings placed inside the box insured a minimum loss of material through the perforated bottom. The boxes were then filled with undisturbed forest floor materials to determine the rate and extent of penetration of this radionuclide into the forest floor and underlying 2.5-cm layer of soil. Ten pairs of sample units were located at randomly picked stations within ten 5- by 10-m sections of the tagged forest. Each pair consisted of one unit with 2.5 cm of soil + organic forest floor layer and a companion unit with the organic forest floor layer only.

The boxes were sunk snugly into a hole so that the surface of the forest layer inside the box corresponded to the surface layer of the adjoining forest floor. These units were collected biweekly, brought to the laboratory for measuring Cs^{137} concentrations, and returned to the study site as promptly as possible, generally within the same day.

Rainfall and litterfall

Periodic measurements of canopy leaching, tree stem flow, and litter fall were taken to ascertain the influx of Cs^{137} to the surface of the forest floor. Canopy leaching was measured by use of 10 stainless steel funnels (26.7 cm in diameter attached to 4-liter plastic bottles), one placed randomly in each 5- by 10-m square of the tagged forest plot. Tree stem-leaching was estimated by constructing a trough around the trunks of 12 trees approximately 1 m above the ground. A plastic tube was used to divert the intercepted water to a plastic carboy near the base of the tree. To collect litter fall, a 1-bu fruit basket (42 cm in diameter) was randomly placed in each of the 20 5- by 5-m subplots. Collections were made weekly during the litter-fall season (September and October) to minimize leaching of leaves before recovery.

Radioactivity determinations

The Cs^{137} concentrations were determined on the larger sized samples (dm^2 soil samples, forest-floor sample units, and litter-fall samples) using a single channel gamma spectrometer. Each of these units and Cs^{137} standards, with configurations matched closely with those of samples, were counted 2.5 cm above a round 7-cm NaI crystal using a spectrometer adjusted to cover gamma radiation of energies from .57 to .77 Mev.

Smaller volume samples (including small soil samples, root samples, and liquid samples) were placed in 2.5-cm test tubes and counted in a well-type crystal attached to an automatic sample changer-scaler (Packard Auto Gamma-210).

Inventory of Radiocesium for First Growing Season

The distribution of Cs^{137} throughout the primary components of the tree-soil system for certain times during the first growing season is summarized in Fig. 1. Data were converted to microcuries (μc) per square meter to be comparable with remaining soil data of this report. The 467 millicuries, introduced into tree boles within the 500-m^2 study area, were thus equivalent to 934 μc per square meter of ground surface. Later removal of some troughs indicated that less than 0.7% of the total radiocesium introduced into inoculation troughs remained there. The remaining 99+% entered the tree stem through chisel cuts, was dispersed

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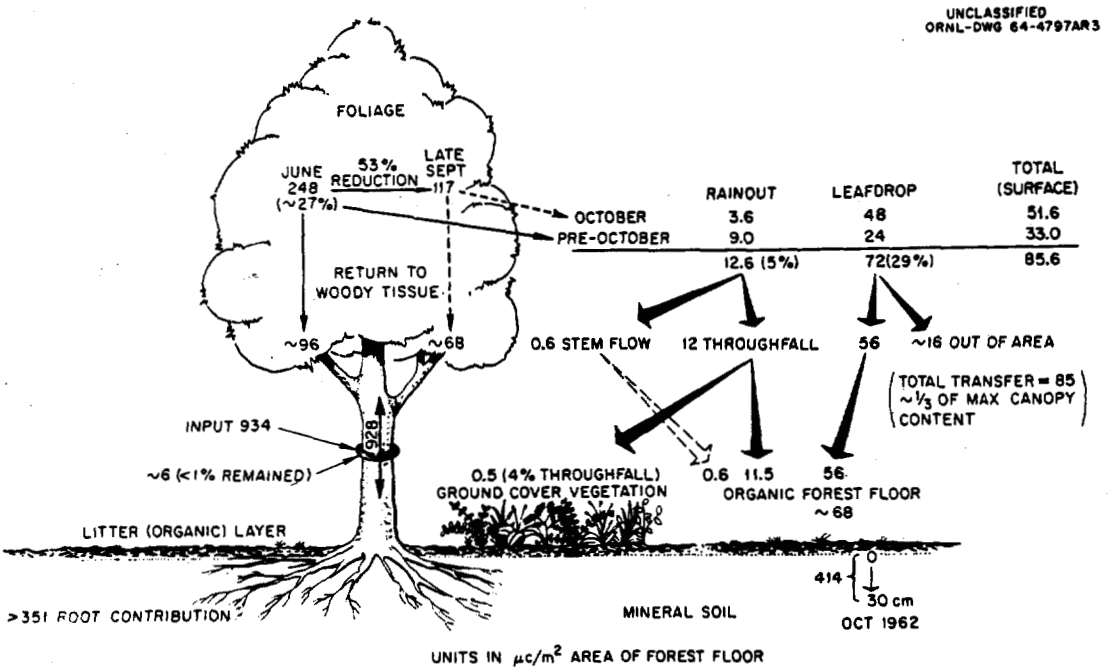

FIG. 1. Flow diagram of transfer of Cs^{137} during the 1962 growing season. Maximum movement into foliage occurred by June (248 μc/m², 26% of total input). During October there was a remobilization of cesium just prior to leaf fall; more than one-half of the cesium in the foliage in late September moved back into woody tissue, and the remainder (about 42%) was transferred to other components of the forest system either by rainout or by leaf fall.

throughout the tree system, and began movement to the surrounding environment.

Forest canopy

Periodic leaf measurements showed that canopy concentration reached a maximum of 27% of the initial input in June (Olson 1965). However, this maximum proportion was reduced to only 12.5% by late September due to several processes discussed below: (1) removal of radiocesium from foliage by rain and dew leaching mostly prior to mid-September which accounted for 5% of the June maximum; (2) 10% reduction by early leaf drop; (3) export of radiocesium from foliage to woody tissue during mature and senescent stages of leaf development, which must account for most of the reduction. Concentrations (μc/g of dry weight) were reduced only slightly by formation of new tissue. Stenlid (1958) summarizes the leaching from mature leaves for other mineral elements, especially potassium, phosphorus, and nitrogen. Such losses from leaves by rain leaching and remobilization and export back into woody tissues have been reported by Long et al. (1956), Middleton (1958), Millar, Turk, and Foth (1958), Tukey, Wittwer, and Tukey (1958), Witherspoon (1964), and others.

Forest floor and soil

Only one-third of the radiocesium found in the forest canopy at peak concentration was transferred to or through the forest floor. Income to the surface of the forest floor area from rain leaching and litter fall of tagged tree components inside the plot was about 68 μc/m².

Most of this Cs^{137} was contained within the organic layer and surface 2.5 cm of mineral soil. Understory vegetation intercepted .5 ± .16 μc/m² out of the total 12.6 μc/m² of Cs^{137} rain leached from the tagged tree canopy. Results from end of the season soil sampling (Oct. 1–15, 1962) indicated a total inventory of 414 μc/m² ± 23 (SE) of Cs^{137} within the 0- to 30-cm mineral soil layer.

Large randomized sample variation.—Our early exploratory sampling indicated a large variation in Cs^{137} concentration between individual samples. Means of four July samples per 5- by 5-m subplot (old litter plus 0–10 cm) ranged from 13 μc/m² in the southeast corner to 141 near the center (Olson 1965, Table 3). Corresponding standard deviations were 8 and 52 for these subplots, as a result of the very skewed distribution of individual samples and plot means around the grand July means of 30.5 μc/m². Differences within and between plots depended on

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TABLE I. Soil-sampling methods and summary of Cs^{137} in organic and soil layers ($\mu c/m^2 \pm$ standard error of mean)

Method 1[a] 200 soil cores (2-cm diameter) in 5-cm sections to 30 cm., including roots but not fresh litter layer		Method 2[b] 20 decimeter squares for finer distribution by 2.5-cm layers and components, including total litter		Method 3[c] Decimeter-square sample slabs of organic layers alone, or organic layers and ~2.5 cm soil	
Organic layers omitted		Organic layers (L and coarse F)	16 ± 2.1	Organic layer box	32 ± 6.90
Mineral soil and roots 0-5 cm	161 ± 10.5	Mineral soil and roots 0-2.5 cm (+ fine F?)	94 ± 10.8	0-2.5 cm soil slab	20
		2.5-5 cm	82 ± 35.6		
5-10	106 ± 9.8	5-7.5	53 ± 15.1	Total retained = (organic + soil) box	52 ± 15.2
		7.5-10	48 ± 21.2		
0-10 Subtotal	267	0-10 Total	277		
		Org. + 0-10 Total	293		
10-15	64 ± 9.6				
15-20	36 ± 5.2				
20-25	27 ± 2.6				
25-30	20 ± 2.4				

[a]Because it is based on a much larger number of samples, method 1 is used for plot totals and average of 267 $\mu c/m^3$ for the 0- to 10-cm layer, instead of 277 based on method 2.
[b]Subdivisions in 2.5-cm layers and various root and soil components from method 2 should be applicable for proportional estimation of the components for the plot mean.
[c]The higher retention of cesium in organic material in the box of method 3 than in the intact forest floor of method 2 might be due to incomplete separation of the finest organic material from the 0- to 2.5-cm layer in method 2 or to a real delay in downward percolation of cesium due to the screen separation of organic layers from the underlying soil.

the distribution of roots from Cs^{137} tagged trees. Witkamp and Frank (1964) also reported that concentrations in 1-cm^3 cores taken along the same transect varied by two orders of magnitude at distances less than 10 cm apart. A high correlation (.94) existed between total activity in a given soil core and activity in roots removed from that soil sample. The results from the randomized sampling were used to describe the average distribution in spite of the large variance. The results of all three methods are summarized in Table I.

Mean soil activity (including roots) at 0–1 dm was 267 in early October, a striking increase from the 30.5 in mid-July as noted above. The standard deviation among 200 individual cores was 243, making the coefficient of variation 91%. Yet, because so many samples were taken, the standard error around the mean was only ± 18.

For the 1- to 2- and 2- to 3-dm segments of the cores, coefficients of variation were still higher (157% and 112%), presumably because the sporadic chance of including or not including roots was almost completely responsible for contributing to the radioactivity of these layers. The mean activities are lower than in the surface decimeter, and standard errors are correspondingly lower: 100 ± 11 and 46 ± 4, respectively.

The depth profiles in successively deeper 5-cm layers averaged 161, 106, 64, 36, 27, and 20 $\mu c/m^2$ in October cores (Table I). In view of earlier results on high root activities and further details given below, the 5- to 30-cm contributions can be interpreted as resulting primarily from a root density gradient ranging from maximum at top and decreasing with profile depth. The 0- to 5-cm layer had still higher root contributions plus additional input from the surface.

The gross spatial variations of radiocesium concentration within the total profile fitted a reasonable pattern relative to the chance distribution of tagged trees. Subplot means ranged from 125 and 169 $\mu c/m^2$ in two 5- by 5-m edge subplots with no tagged trees to 672 $\mu c/m^2$ (northwest corner) and 742 $\mu c/m^2$ (middle of south edge of the plot). The coefficients of variation of profile totals within subplots ranged from 40% to 109% of the plot means. Square root and logarithmic transformations helped to normalize the data and confirmed that the intensive sampling provided estimates within a few per cent of the radiocesium content, in spite of the high variance and skewed distribution (Fig. 2).

Detailed soil component sampling.—With the roots apparently contributing such a high percentage of Cs^{137} activity to the soil system, it became

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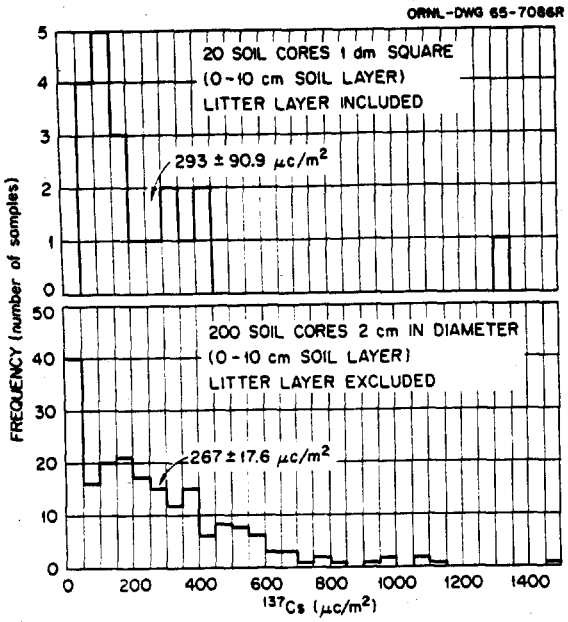

FIG. 2. Histogram depicting variation of Cs^{137} between individual samples. Top: 1-dm-square area samples used to determine distribution of radiocesium between forest floor components. Bottom: 2-cm diameter cores used to determine the total budget of radiocesium within the forest floor.

desirable to associate the radionuclide with the different components of the soil. Twenty decimeter-square area samples were taken to provide a larger volume of material for inclusion of larger roots and stone than could be sampled in cores. Results indicate a total burden of 293 ± 90 μc/m² in the litter layer plus 0- to 10-cm soil layer. The smaller number of samples using this method resulted in a 30% standard error around the mean, chiefly because of one outlying value (Fig. 2, top). Nevertheless, the mean for mineral layers (277 μc/m²) compares surprisingly well with the mean of 267 from the 200 core samples (Fig. 2).

These sampling techniques give a minimum estimate of the Cs^{137} within the organic forest floor layers. An attempt was made to separate the organic L and F layers from the mineral soil layer, but it was not always feasible to separate the fine fragmented F materials from the upper mull humus soil layer. Therefore, the decimeter cube samples (indicating a 16 μc/m² burden for the organic L and coarse F layers) may underestimate the total organic layer burden. Witkamp and Frank (1964) reported that the older part of the F layer retained considerable Cs^{137} that was leached from the recently fallen leaves.

Forest floor boxes.—The decimeter-square plastic boxes probably gave a maximum estimate of 32 μc/m² within the organic forest floor (L and F layers—Table I) and showed changes through time. The forest floor layer was placed inside these boxes at the beginning of the experiment, and the litter fragments from decay of L and F layers throughout the season were contained largely within these sample units.

The surface mineral soil layer was retained inside half of the plastic boxes. Since roots were kept out, accumulation corresponded closely with amounts of surface income from rain leaching of Cs^{137} from tagged leaves and from fallen litter. The bar graph on Fig. 4 compares the seasonal changes of such income with the accumulation of Cs^{137} in the surface 2.5 cm of mineral soil contained within plastic boxes.

Loss Out of Area

Leaf drop

Measurements at the end of the second growing season (October 1963) provided estimates of movement of Cs^{137} outside the 20- by 25-m study area by falling of tagged leaves. Results from 174 samples (.1 m area each) collected at 5-m intervals along transects extending 20 m beyond the east and west boundary of the study area and 10 m beyond the north and south boundary, indicated that 2.67 mc (equivalent to 5.34 μc/m² area of study site or 0.57% of the total radiocesium) was transported outside the study area by leaves from tagged trees during the second growing season. This pattern should not differ greatly from year to year because of the protected location of the forest, but the amount of radiocesium exported should have been greater in 1962 than in 1963 since leaf concentrations were higher in the former year.

Foliage concentration just prior to leaf fall averaged 0.6 μc/g of leaves in 1962 and 0.2 μc/g in 1963. Therefore, Cs^{137} deposited outside the study area through leaf fall for the first growing season (1962) should have been about three times the 5.3 μc/m² removed through leaf fall at the end of the second growing season (1963). This estimate of 16 μc/m² is a significant amount when compared with the 55 μc/m² that was estimated in collections of litter inside the experimental study plot and suggested that about one-fifth of the leaves from the tagged trees fall outside the delineated 20- by 25-m area. However, more than three-quarters of this activity is located within a 10-m zone around the perimeter of the study area.

Surface runoff

Lateral movement of radiocesium out of the area due to surface runoff was apparently small. Soil

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samples taken at 1-m intervals for a distance of 3 m from the study plot were not statistically different from control samples unless a root from a tagged tree was intercepted.

Surface Income to the Forest Floor

The main components of surface income to the forest floor included litter fall and rain leaching of the aerial parts of the tagged trees. The contributions of herbivores (especially larvae of Lepidoptera), which are active in transferring nutrients, were not separated from that of litter fall. Foliage-feeding insects may have removed a small percentage of the leaf area in June, but during the remainder of the season their activity diminished.

Rainfall

Rainfall leaching of the forest canopy accounted for about one-fifth of the total surface income and was the major process transferring this radionuclide to the untagged ground-cover plants (Fig. 1). Rain falling through the canopy accounted for 12 ± 2.5 μc/m². Radiocesium flowing down tree stems was distributed very unevenly, but it was equivalent to an average of only 0.6 μc/m² of soil area. Although stem flow is one order of magnitude more concentrated per milliliter of rain than canopy rain-through (Fig. 3), radiocesium collected from tree stems accounted for only 5% of the total leached from the tagged trees. Stem flow is undoubtedly very important in the nutrient cycles of lichens and mosses growing on or near the base of the tagged trees and contributes to the high variance of soil radioactivity.

The rate of leaching to the forest floor was greatest in early June when the canopy concentration was highest, and increased again in September during the senescent stage of leaf development (Fig. 4). Throughout the early months, Cs^{137} leached out of the canopy contributed more than 80% of the incoming Cs^{137} flux (Fig. 5). By late August, both rainout and leaf drop had each contributed about 6 μc/m². Cumulative rain-

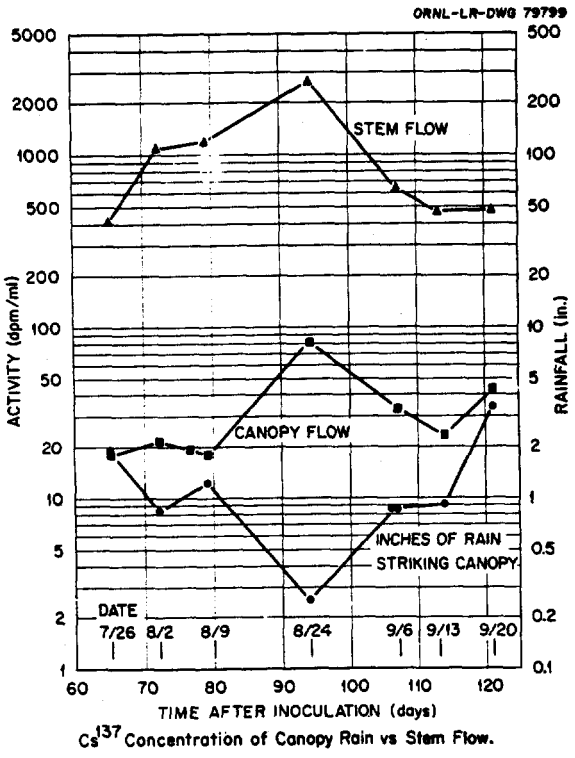

FIG. 3. Cesium-137 concentration of canopy rain vs. stem flow.

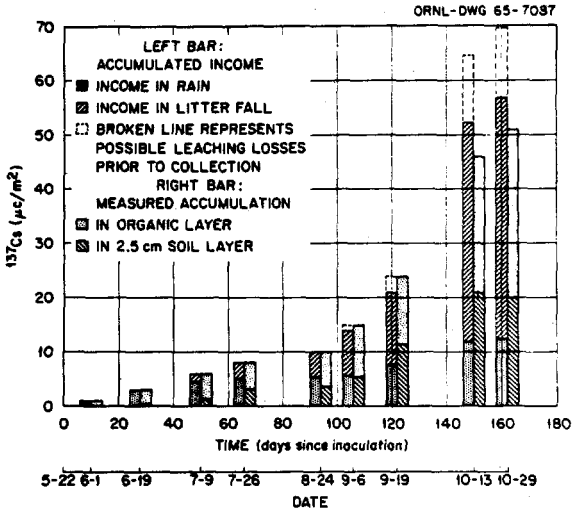

FIG. 4. Cumulative surface income and retention of cesium-137 within the surface layers of the forest floor.

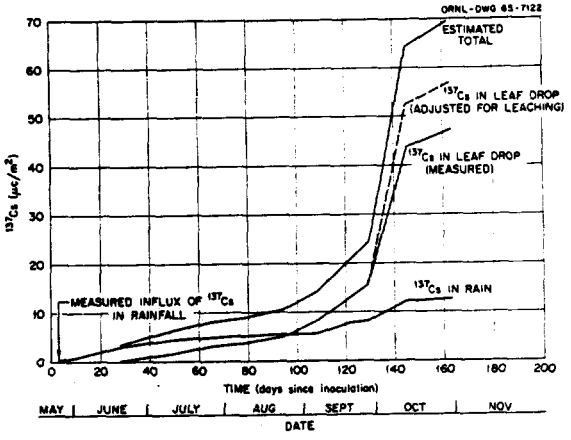

FIG. 5. Total cumulative income of Cs^{137} to the surface of the forest floor.

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TABLE II. Cesium-137 distribution by forest-floor component to a depth of 10 cm in mineral soil ($\mu c/m^2$ ground area)

Component	Mean	± 1 SE	SD	Coefficient of variation (%)
Vegetation[a]	0.5	.16		
Litter layer[b] (does not include fine material)	15.7	2.1	9.5	60
Total organic layer[c] (includes fine material)	32.5	7.4	21.0	65
Rock (> 2mm)[d]	9.0	2.6	11.8	131
Medium root[e] (1-15 mm)	56.6	24.0	106.9	189
Fine roots[f] (< 1 mm)	27.6	1.8	4.1	15
Soil including fine roots and organic materials	211	64	287	136

[a]20 harvested samples of vegetation (.1 m area each).
[b]20 dm^2 samples-intact leaves and large leaf fragments from surface.
[c]10 dm^2 samples-units encased in plastic.
[d]20 dm^3 samples-greater than 2 mm portion.
[e]20 dm^3 samples-root fragments hand separated.
[f]Fine root fragments separated from 20 subsamples (50 g each) by flotation.

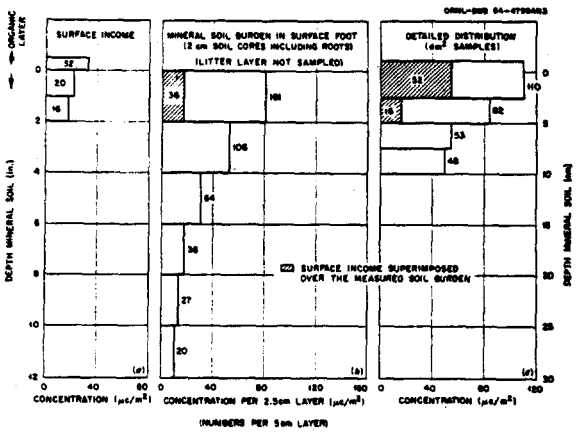

FIG. 6. Vertical distribution of Cs^{137} within forest floor. Litter separated in (a), not sampled in (b), and grouped with the 0- to 2.5-cm layer of soil in (c).

out had doubled by late October, but was relatively less important than leaf drop in autumn.

Leaf drop

Over two-thirds of the litter fall contribution occurred during the 30-day period between mid-September and mid-October (Fig. 4, 5). Although fallen leaves were collected at weekly intervals during periods of greatest leaf fall, results from litter bag studies indicated that as much as 30% of the Cs^{137} may be released from a freshly fallen leaf during the first week. To indicate an upper limit of the compensation for leaching losses prior to collection, the measured activity of 47 $\mu c/m^2$ in litter fall has been increased 20% to 57 $\mu c/m^2$ in dashed upper line of Fig. 5. This adjusted estimate was shown in Fig. 4 and compares favorably with the estimated income that was measured by the decimeter-square forest-floor and soil-sample units until late September (Fig. 4). Possible underestimates of leaf-drop contribution should be approximately compensated for as overestimates in the rainout contribution, due to the leaching of leaves that were caught in the rainfall funnels between collections. Therefore, our upward adjustments of total surface income may, if anything, be excessive.

Even with generous allowance for surface income to the forest floor, the estimated cumulative total of about 68 $\mu c/m^2$ is far less than the 414 $\mu c/m^2$ that needs to be accounted for in the soil inventory. Furthermore, the following details on the distribution of Cs^{137} introduced at the ground surface will confirm that this contribution was retained relatively close to the surface, and that it failed to account for the inventory of radioactivity below the surface decimeter shown in Table I.

Accumulation of surface income

The forest-floor sample units in plastic boxes indicated 32 $\mu c/m^2$ of this surface income was retained within the organic floor layer (Fig. 6a). The sample units containing the organic layer plus 2.5 cm of mineral soil contained 52 $\mu c/m^2$ of Cs^{137}. Fig. 6a suggests that about 16 $\mu c/m^2$ of the total 68 $\mu c/m^2$ were left to penetrate to depths greater than 25 cm within the mineral soil layer.

Much of the total surface income was thus temporarily immobilized within the organic litter layer (Fig. 4). Witkamp and Frank (1964) reported that immobilizations of radiocesium occurred within the older decomposed litter layer in the forest and in laboratory cultures. They suggest that soil microorganisms contributed to the immobilization since some mushrooms were found to have concentrated Cs^{137} 80 times over the concentrations of the substrate. Barber and Mitchell (1963) pointed out the ability of organic materials to immobilize Cs^{137} enough to prevent transfer by percolating water.

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Subsurface Income: Roots

About half of the total Cs^{137} inoculant was within the forest soil system only 5 months after inoculation. Roots of the tagged trees must have contributed much more Cs^{137} than leaching from leaves and litterfall. The total Cs^{137} burden of 414 ± 23 $\mu c/m^2$ in the surface 30-cm soil layer greatly exceeded the surface income reaching the mineral soil layer (about 36 $\mu c/m^2$, Fig. 6a, 6b).

Distribution

Since roots contributed a high percentage of activity to the soil system, an attempt was made to determine the root contribution and the distribution of Cs^{137} between the forest soil components. The decimeter samples of forest soil were divided into six components; the results of manual, flotation, and chemical separation are given in Table II.

Manual separation.—This separation (Fig. 7) indicated that medium-sized roots (1–15 mm) contributed almost as much activity as all surface income combined:

Surface income (litterfall + rainleaching — Fig. 7)	68 $\mu c/m^2$
Subsurface income (> 1 mm roots from tagged trees — Table II)	57 $\mu c/m^2$
Total	125 $\mu c/m^2$

Then subtracting the above total (accountable income) from the total burden of 293 $\mu c/m^2$ for the decimeter square samples leaves a deficit of 183 $\mu c/m^2$.

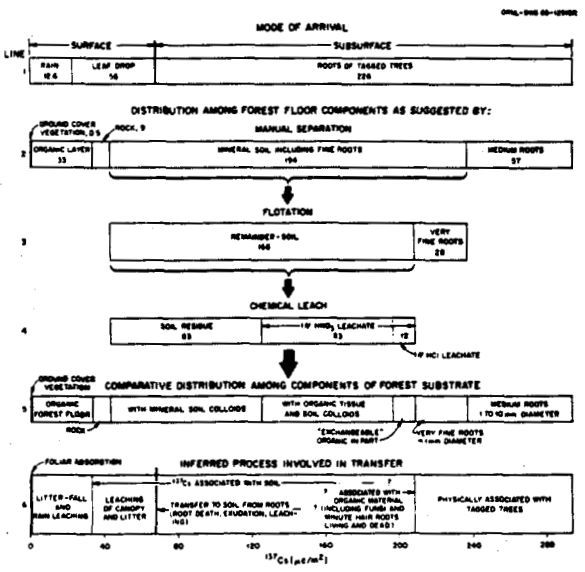

FIG. 7. Flow diagram and summary interpretation of Cs^{137} in components of decimeter-square forest floor soil samples, 0-1 dm deep ($\mu c/m^2$).

Flotation separation.—A further separation of soil components was made by using difference in specific gravity between mineral soil and fine organic material (Fig. 7, line 3). The fine organic material (including < 1 mm roots) was removed and concentration determined and corrected for Cs^{137} activity measured in water. Results of manual and flotation separations are:

Surface income (litterfall + rain leaching — Fig. 1)	68 $\mu c/m^2$
Subsurface income	
(> 1 mm roots — Table II)	57 $\mu c/m^2$
(< 1 mm roots — Table II)	28 $\mu c/m^2$
Total	153 $\mu c/m^2$

Surface income and Cs^{137} still retained in roots accounted for only about one-half of the total 293 $\mu c/m^2$, so a further separation was done on the soil component.

Chemical separation.—Three leaching reagents took advantage of the difference in ionic bonding or physical entrapment of Cs^{137} in the living and dead components of the above soil: tap water, 1 N hydrochloric acid (nonoxidizing), and 1 N nitric acid (oxidizing acid). The percentages of Cs^{137} removed with the three reagents were as follows: tap water, 0.3 ± .04; 1 N HCl, 7.4 ± 1.0; and 1 N HNO_3, 50.6 ± 6.9.

There are two nearly equal cesium-137 pools within the mineral soil layer after the flotation separation: (1) the half remaining with the soil residue after the nitric acid treatment surely represents Cs^{137} associated with mineral soil; (2) the half removed by this treatment presumably represents Cs^{137} associated with mineral soil (but not bound so firmly as the preceding pool) and Cs^{137} associated with organic materials (Fig. 7, lines 4 and 5).

Importance of roots for radiocesium income

An analysis of the radiocesium income to the forest floor and its redistribution indicates quantitatively how important roots are for transfer back to soil (Fig. 7, line 6). Of the total activity found within the mineral soil layer (277 $\mu c/m^2$ including roots and rock) we could physically associate less than 50% directly with the tagged trees themselves at the time of soil collection. Presumably many ions entered the soil by contact exchange, to maintain the electrical balance of roots during absorption by nutrients. Much of the remainder may be accounted for by Cs^{137} entering the soil

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system by root exudation, die-off of small roots and root hairs, transfer to mycorhizal fungi, and leaching of roots by the soil solution. All of these processes are known to occur, but their quantitative or relative importance in transfer of materials to mineral soil is not very well documented.

For this reason the early tagging results are of special interest for evaluating the relative importance of belowground and aboveground transfer of mineral elements. If that portion of Cs^{137} remaining in the mineral soil residue after the nitric acid treatment (Fig. 7, line 6) is considered to be the only fraction completely disassociated from roots at the time of sampling, then the subsurface transfer of radiocesium out of the roots is about as great as the aboveground transfer. However, later nitric acid leaching experiments with Cs^{137}-spiked soils suggest that a major portion of the Cs^{137} associated with the nitric acid leachate (Fig. 7, lines 4, 5) was also associated with mineral soil colloids. This would indicate that the turnover of Cs^{137} (and probably other alkali metals) from tagged yellow poplar trees is greater belowground than aboveground.

Although the promptness of underground turnover just described appears surprising at first, the possible importance of such transfer has been suggested by several workers and possible explanations for transfer have been offered. Stenlid (1958) and Fujiwara and Iida (1956) discuss possible losses of materials from roots due to excretion and leaching. Based on estimates of Orlov (1955), Remezov (1958, 1959) emphasized the possible mineral and organic turnover due to production and death of roots throughout the season. He estimated that .4 metric tons per hectare, half of the amount produced, died per year in a 50-year-old oak stand. He also indicated that the death of small roots < 0.3 mm in diameter with high mineral content affects the mineral cycle significantly. Although they did not attempt a quantitative budget, Woods and Brock (1964) inferred that root transfer of minerals may be greater than is normally thought, and they cited further papers on the role of root exudates and mycorrhizal fungi as avenues of transfer to the rhizosphere and other species.

Discussion and Conclusions

An inventory of total Cs^{137} distribution among the tree and soil compartments of a tulip poplar forest (tagged May 1962) was constructed from data collected during the first growing season. A measurable influx of the radiocesium to the ground was recorded only 3 days after inoculation and over 40% of the initial inoculant was found in the litter, soil, and smaller root components by October (Fig. 1). The maximum burden found in the canopy about 4 weeks after inoculation (248 $\mu c/m^2$ of ground area) was reduced to 117 $\mu c/m^2$ by late September. Of the 131 $\mu c/m^2$ reduction, export of Cs^{137} from foliage to woody tissue by mature and senescent leaves accounts for almost 75%, rain and dew leaching about 7%, and early drop of leaves 18%. October losses were about 68, 3.6, and 49 $\mu c/m^2$ by translocation, rainout, and leaf drop respectively.

The important paths of transfer of cesium-137 from tagged trees to the forest floor included not only rain leaching and leaf fall, but especially passage through the roots. Transfer to the untagged understory vegetation by foliar interception of radiocesium leached from overhanging crowns of tagged trees is initially far more important than uptake from Cs^{137} in the soil (Waller, Olson, and Cristofolini 1965).

This study, like a previous one using small white oak (*Quercus alba*) trees (Witherspoon 1964), thus provides quantitative measures confirming the rapid translocation of Cs^{137} throughout major plant components of a forest. By contrast, cesium is a relatively immobile element when in direct association with the soil and tends to accumulate there (cf. Nishita, Steen, and Larson 1958, Titlyanova and Timofeyeva 1959, Schultz, Overstreet, and Barshad 1960, Lai, Nuang, and Chang 1961).

Woods and Brock (1964) concluded that the rate of transfer from roots to soil was much faster for P^{32} than for Ca^{45}. Wittwer, Bukovac, and Tukey (1963) listed comparative basipetal movement within plants from nutrients applied on foliage in four categories: (1) highly mobile elements —nitrogen and alkali metals; (2) mobile elements—phosphorus, chlorine, and sulphur; (3) partially mobile—zinc, copper, manganese, iron, and molybdenum; and (4) immobile—boron and alkaline earths.

If this relationship between basipetal mobility within plants is similar to that of transfer *from* root systems, then one would expect the other members of the alkali metals to be transferred at high rates relative to the members of the alkaline earth group such as calcium and magnesium. Alkaline earth elements should be among the least mobile elements in their contribution to soil by downward movement through plants, but they should have a relatively greater contribution to the cycle through surface income including leaching and leaf fall of the aerial components.

If such regularities in transfer rates apply widely

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among element groups in natural vegetation, this should be helpful in drawing broader nutritional interpretations from cycling studies using non-nutrient elements like cesium and strontium which have convenient radioactive isotopes for tracer research. Generally alkali metal elements are consistently among the most rapid in cycling within the organic parts of ecosystems, even though many are readily immobilzed after intimate contact with soil minerals.

The use of isotopic Cs^{137} as a tracer in trees may have other practical applications such as defining the biomass of roots, especially the very fine roots that are very difficult to separate from soil and even more difficult to separate from the roots of herbaceous or ground-cover vegetation. If one can ascertain typical concentrations of Cs^{137} per unit weight of different root sizes prior to any great release of this radionuclide from the tagged tree, then an estimate of root mass simply becomes one of soil-root sampling and radioassaying without the tedious task of root separation. Also, the task of describing the total soil volume occupied by a tree root system, especially those species with a small frequency of root grafts, could become less laborious. These possibilities can extend the already long list of uses of radionuclides to help interpret underground phenomena associated with roots.

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