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RADIOCESIUM CYCLING IN VEGETATION AND SOIL

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ABSTRACT

Data on cesium dynamics in vegetation and soils are reviewed. Special emphasis is placed on typical environments in the southeastern United States. Clay minerals of soil, especially micaceous types, effectively fix cesium and remove it from biotic components of ecosystems. Fallout ^{137}Cs enters food chains principally by direct deposition on vegetation; uptake by the root pathway is usually less than direct contamination of plant foliage. Cesium-137 levels in vegetation of the southeastern U. S. Coastal Plain are estimated on the basis of direct deposition from the atmosphere and root uptake from soil. Estimated concentrations, based on current concepts of ^{137}Cs dynamics in vegetation and fixation in soil, are in good agreement with observed values of ^{137}Cs in vegetation collected in 1969 and 1970. Mechanisms of direct deposition and of increased uptake by roots due to the absence of micaceous clays adequately explain the higher levels of ^{137}Cs in vegetation of the Coastal Plain and thus are responsible for the elevated ^{137}Cs reported for milk of the Tampa, Fla., milkshed.

Considerable interest has been shown in the dynamics, fate, and effect of ^{137}Cs in the biosphere because it is a predominant long-lived product in the fission of nuclear explosives and reactors. Approximately 6% of the total fission of ^{235}U is ^{137}Cs. Cesium-137 has been widely distributed as global fallout; low levels occur in all parts of the biosphere. Because it is readily assimilated into body tissues, ^{137}Cs becomes a major contributor to radiation dose to living organisms.

Adequate technology currently exists in the nuclear fuel cycle to control releases so that ^{137}Cs from nuclear waste will not exceed the current levels caused by fallout from nuclear explosions in the atmosphere. Unless atmospheric testing resumes, we would not expect to again experience elevated environmen-

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tal concentrations of ^{137}Cs from fallout. Indeed, the peak level of ^{137}Cs in the world atmosphere and biotic components of ecosystems probably has already occurred according to monitoring data obtained for the past 25 years (Radiological Health Data and Reports 1963–1973; HASL 1974). The reasons are threefold: (1) in the absence of new ^{137}Cs injection to the atmosphere, the deposited quantities are diminishing owing to physical radioactive decay; (2) experimental data and fallout monitoring results show that the greatest ^{137}Cs assimilation into biological organisms is coincident with the initial ^{137}Cs introduction into the ecosystem, i.e., direct contamination of vegetation followed by ingestion of foodstuffs by livestock or humans; (3) cesium is effectively removed from biological circulation through the mechanism of fixation by soil clays.

In this paper we review the factors that influence the cycling of cesium in terrestrial ecosystems. The review focuses on the mechanisms influencing the dynamics and fate of ^{137}Cs in pathways leading to man. A limited amount of new data confirms existing concepts of cesium cycling in biota and retention in soils. Existing knowledge of ^{137}Cs dynamics is applied to the interpretation of elevated ^{137}Cs in milk in the Tampa, Fla., milkshed.

GEOCHEMISTRY OF STABLE CESIUM

In a mineral cycling symposium (actually a misnomer because it is elements rather than minerals that cycle), it would be instructive to review the natural geochemistry of stable cesium before addressing the cycling of ^{137}Cs in biotic and abiotic components of ecosystems. The basic geochemistry of cesium provides substantial insight into the possible fate of fission-product ^{137}Cs in geological substrates. The primary minerals that contain cesium are pollucite, rhodizite, and avogadrite (Fairbridge, 1972). Cesium is isomorphically substituted for potassium and rubidium in the lattice of other minerals, e.g., cesium for potassium in the formation of biotite and leucite. Biotite can also serve as a secondary source for cesium. As the crystalline structure is decomposed in processes of mineral weathering, cesium is solubilized and immediately resorbed to clays.

There is ample geologic evidence supporting the concept of interlayer lattice fixation of cesium because only in those sedimentary deposits which contain clays or shales is the cesium abundance greater than 1 ppm. In contrast, the concentration level is in the parts per billion range in sandstone and limestone. Cesium ions are adsorbed to selective sites on clays of geologic sediments or they penetrate into interlayer voids of micaceous minerals and, for all practical purposes, are permanently fixed. Thus, beginning with a primary mineral, the sequence of events in the geochemistry of stable cesium is: mineralization from primary minerals → substitution in micaceous minerals → release in subsequent weathering processes → selective fixation (or adsorption) by clay minerals.

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REACTION OF ^{137}Cs WITH CLAY MINERALS

Research in recent decades has confirmed the mechanism for fixation of ^{137}Cs by clay minerals (Shulz, Overstreet, and Barshad, 1960; Tamura and Jacobs, 1960). This mechanism is by far the most important factor in the biogeochemical cycling of cesium in terrestrial ecosystems because the degree of fixation by clays governs subsequent availability to biological organisms. In addition to adsorption by the general cation-exchange complex of soils, cesium is effectively adsorbed to selective sites, and fixation occurs with weathered potassium-depleted micaceous clays. Lomenick and Tamura (1965) reported that fixation varied with different clay minerals. With micaceous-type minerals and with clays exhibiting a limited expanding lattice crystalline structure, desorption of ^{137}Cs (the inverse of fixation) was 30% of that for nonexpanding kaolinitic lattice type and fully expanding montmorillonite type minerals. The latter minerals (kaolinite and montmorillonite) are known for their nonselective cation-exchange reactions, whereas the former (mica and illite) exhibit high selectivity.

The mechanism for fixation of ^{137}Cs is related to the weathering of potassium from micaceous clays. When added to soil, cations whose radii are similar to that of potassium, such as cesium, rubidium and NH_4^+, are tightly adsorbed to these weathered micaceous minerals. If sufficient quantities of these cations are added, the expanded lattice collapses because of the favorable ionic radii of these elements and because of the neutralization of the negative charges between the clay surfaces, and cations are fixed or trapped (Arnold, 1960; Barshad, 1954). This mechanism likely accounts for fixation of potassium, cesium, rubidium, and NH_4^+ cations when added to soil in macroquantities (>10 ppm).

Different mechanisms likely prevail in the fixation of microquantities (ppb range). For example, illite and micas, as well as collapsed vermiculite (potassium saturated) and bentonite (heat treated), strongly adsorb trace levels of ^{137}Cs (Tamura and Jacobs, 1960). Fixation is considered to be largely near the edges of the interlayer spacings. Density-gradient zonal-centrifugation techniques have been used to identify clay-mineral adsorption sites of ^{137}Cs in soil (Francis and Tamura, 1972; Auerbach, Olson, and Waller, 1964) collected from the ^{137}Cs tagged *Liriodendron* forest. The dominant mineral responsible for ^{137}Cs adsorption in this soil was pedogenic chlorite (a mineral derived from the weathering of mica).

Results on ^{137}Cs desorption from soils with different predominant clay minerals confirm that fixation is greatest with micaceous clays. Tracer quantities of ^{137}Cs (carrier-free solution) fixed to three podzolic soils from Tennessee were desorbed with 1*N* sodium acetate (Table 1). In the initial 0.25 hr, 32% of the ^{137}Cs was desorbed from the kaolinitic Fullerton soil, and desorption continued for 264 hr (87%). The illitic Sequoia soil showed maximum desorption (about 50%) in about 1 to 4 hr; then the soil appeared to resorb the

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TABLE 1

PERCENTAGE DESORPTION OF RADIOCESIUM FROM THREE EAST TENNESSEE SOILS (A HORIZON)

Soil	Solution contact time, hr*					
	0.25	1	4	24	96	264
Fullerton (kaolinitic)	32.2	46.2	59.3	75.3	80.6	86.7
Sequoia (illitic)	47.8	50.8	49.0	37.1		17.7
Captina (mixed)	27.0	33.4	36.5	33.7	32.3	29.4

*Desorbing solution was 1*N* sodium acetate; 100 ml of solution per gram of soil; maximum desorption is underlined.

cesium, resulting in 18% desorption after 264 hr. Thus, in 1*N* sodium acetate, desorption of ^{137}Cs was a factor of almost 5 greater with kaolinitic soil compared with that for illitic soil. The Captina soil contains both kaolinite and illite in about equal quantities; its desorptive characteristic resembles that of the illitic Sequoia.

The desorption data (Table 1) provide further insight into the role of selectivity of adsorption sites in the fixation of ^{137}Cs by various clays. The adsorption of cesium occurred from a distilled-water system, and the trace level of cesium likely was adsorbed on surfaces with which it came in immediate contact without regard to selective or nonselective sites. When sodium ions were introduced during desorption, the cesium ions could be displaced from nonselective sites; the quantity displaced increased with time of contact. This type of response is typified by the kaolinite-bearing Fullerton soil. On the other hand, with the illite-bearing Sequoia and Captina soils, the desorbed cesium from nonselective sites was readsorbed on illitic selective sites (Tamura and Jacobs, 1960). Sodium ions can desorb cesium from the nonselective sites but cannot compete with cesium for the selective sites; hence the decreasing desorption observed in the illite-bearing soils represents readsorption at the selective sites.

Minerals and clays possessing selective sites (vermiculite: Sawhney, 1965; illite and hydrous mica: Tamura, 1964) have high selectivity for cesium. Francis and Brinkley (1975) provided additional information on the role of minerals and clays in the fixation of ^{137}Cs in sediments from Steel Creek below the AEC Savannah River Plant in South Carolina. The concentration of ^{137}Cs in the micaceous component was three times that of the kaolinite (3.6 to 1.1 nCi/g, respectively). Although concentration of ^{137}Cs was greatest on the micaceous clays in this sample, over 90% of the ^{137}Cs was associated with the kaolinite mineral simply because this mineral is the predominant clay in soils and sediments of the Savannah River region.

Fixation of radiocesium by soil is the major factor that determines the long-term availability to biological organisms in terrestrial ecosystems. Cesium

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removal from biological circulation in ecosystems is mediated by soil clay and in particular by such cesium-fixing clay minerals as illite. Other micaceous-derived clays (vermiculite, pedogenic chlorite) also selectively adsorb cesium. Thus, in soils where these minerals predominate or are even found, the uptake of radiocesium by plants is severely restricted because the soil serves as an effective sink for fallout ^{137}Cs.

CESIUM-137 TRANSPORT IN TERRESTRIAL ECOSYSTEMS

Transport of cesium in terrestrial ecosystems involves both physical processes (by atmospheric dispersion, aerosol dynamics and deposition, by movement in hydrologic streams, and by erosion of sediments) and biological processes (assimilation of foliar deposits, root uptake, and food-chain transport). Following its incorporation into the vegetation component, the ^{137}Cs is dispersed among naturally occurring food webs. For selected food-chain pathways, the nuclide can be ingested by man. Direct deposition on vegetation and immediate incorporation into food chains dominates initial phases of biological transport. In the absence of new ^{137}Cs from atmospheric deposition, the soil–plant pathway constitutes the principal mode of ^{137}Cs transport to man.

Transport of cesium to man can be many faceted (see Fig. 1 in Kaye and Ball, 1969), but the principal pathways for ^{137}Cs are those described by Bruce and Russell (Fig. 1 in Bruce and Russell, 1969). For food-chain mechanisms Bruce and Russell conclude, "a relatively small number of critical pathways are responsible for . . . airborne radioactivity that reaches man's diet." They also conclude that the most direct route is direct foliar contamination → milk → man for exposures involving ^{137}Cs. In any case, the hazard to man from ^{137}Cs will involve these transfers; direct deposition on foliage, soil to plant, plant to animal or man. Emphasis on only these transfers admittedly is an over simplification, but it is beyond the scope of this paper to describe the innumerable pathways for both agricultural and ecological systems.

Direct Foliar Contamination

More than a decade ago Russell (1963, 1965) recognized that direct foliar contamination rather than uptake from soil was the principal mode of entry of fallout fission-product nuclides into vegetation. Others (Krieger, Kahn, and Cummings, 1967) have shown that uptake of ^{137}Cs from soil accounts for only 3% of the total burden in bluegrass, the remainder being contributed by direct deposition (Table 2).

Monitoring data illustrate the close relationship between ^{137}Cs concentration in air and ^{137}Cs in milk. A significant positive correlation ($r = 0.97$) for ^{137}Cs concentration in air and milk was observed for quarterly averages from seven milksheds across the United States from 1962 to 1967 (Wilson, Ward, and

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TABLE 2

COMPUTED RELATIVE CONTRIBUTIONS OF ^{137}Cs TO BLUEGRASS FROM FALLOUT CONTAMINATION OF FOLIAGE AND FROM ROOT UPTAKE OF CONTAMINATED SOIL*

Basis	Deposition on foliage, pCi/m²	Root uptake, pCi/m²	Ratio root uptake / total
Stable cesium†	210	6	0.03
Uptake of ^{137}Cs from soil	207	8	0.04
Covered plots	154	62	0.29

*Data from Krieger, Kahn, and Cummings (1967), p. 69.

†Root uptake calculated on basis of stable cesium concentrations in vegetation and soil.

Johnson, 1969). A multiple regression model, based on fallout over the preceding 2- to 6-month period, predicted with considerable accuracy the declining concentration and seasonal variation of ^{137}Cs in milk. The high correlation of ^{137}Cs concentration in air and concentration in milk strongly implies that direct deposition on vegetation was responsible for much of the ^{137}Cs entry into the grass–cow–milk food chain. To date, the concentration of ^{137}Cs in air and milk has continued to decline (Fig. 1). The significance of these data is that levels in air and milk have decreased simultaneously by an order of magnitude since the moratorium on nuclear testing in the atmosphere. In contrast, a reverse trend would be expected a priori if concentration in milk were directly related to accumulation of ^{137}Cs in soil, i.e., levels in milk would increase with cumulative ^{137}Cs in soil.

Various investigators have demonstrated that nutrient elements (Biddulph, 1960) and fission products (Middleton, 1959; Levi, 1969) are readily assimilated by foliage. The results show that ^{137}Cs is relatively mobile within this vegetation component. From 10 to 20% of the assimilated radiocesium may be translocated to grain (Middleton, 1959) or fruiting structure (Levi, 1969). The mechanism of entry into leaf-cell protoplasm appears to involve adsorption of cations to negatively charged biocolloids or nondiffusible anions in the cell wall, followed by active metabolic uptake into the cell (Biddulph, 1960). Initial adsorption would be mediated by surface fissures in the cutin which would allow entry of soluble fission products to anionic sorption sites or to plasmodesmic extensions to cuticle margins. However, the exact mechanism and the kinetics of ionic transport from external surfaces to internal protoplast are not known. In practice, however, it may not be essential to know the relative distribution between internal and external plant parts because both external and internal plant structures are consumed by foraging animals. The concentration of

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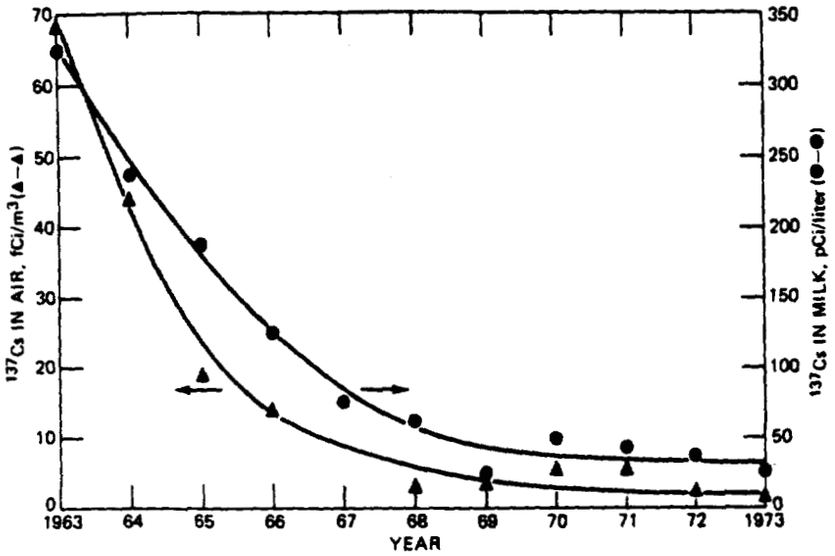

Fig. 1 Parallel relationship between concentration of ^{137}Cs in air (fCi m^{-3}) and in milk (pCi/liter). Close agreement over an order of magnitude decrease in both parameters suggests a strong relationship between direct deposition and entry of ^{137}Cs into food chains. Data taken from USAEC Health and Safety Laboratory Fallout Program (1974) and from Pasteurized Milk Network (1973).

contaminants in foodstuffs (representing both external and internal contamination) is the most important information needed for estimates of trophic transport, and the internal vs. external localization of ^{137}Cs in the leaf may be of secondary importance.

Retention and Weathering of ^{137}Cs from Contaminated Foliage

The processes of retention and weathering determine time-dependent concentrations of the contaminant in vegetation. The fraction of the incident contaminant which is retained by the vegetation depends on the physiochemical form of fallout as well as on the foliar characteristics of vegetation. For grass contaminated by aqueous spray or by fallout particles, representative retention values are given in Table 3. The wide range of initial retention of fallout (aqueous or particulate form) is partially explained by different foliage density, but other factors, e.g., mode of deposition, morphological differences in foliage, and duration between application and measurement, may all influence estimates of initial retention. For example, the effect of leaf morphology is illustrated by the fact that 75% of total retention by sorghum was located in the leaf axil (junction of leaf blade with stem axis) (Witherspoon and Taylor, 1971). Furthermore, substantial losses may occur between the time of deposition and

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initial measurements because only 11% of initial deposition of fallout simulant particles remained on fescue after 1 hr (Dahlman, 1971).

The time-dependent decreases of ^{137}Cs following direct foliar contamination are also somewhat variable. Russell (1965) reports an average weathering half-time of 14 days for vegetation contaminated by natural fallout. We have determined from experiments with ^{137}Cs-tagged fallout simulant applied to fescue grass that decrease in ^{137}Cs content of the grass mimicked a negative exponential, $Y = 0.27 + 1.59e^{-0.52t}$, where Y is retention and t is time in days. For the nonlinear regression fit, the weathering half-time is approximately 30 days. If a negative exponential loss rate is assumed, Middleton's (1959) data for aqueous-spray contamination suggest a 32-day weathering half-time. From different observations it appears, then, that approximately half the ^{137}Cs deposited on foliage is lost from grass vegetation in an interval of 14 to 30 days.

TABLE 3

RETENTION OF FALLOUT BY GRASS

Mode of application	Species	Initial retention, %	Reference
Aqueous spray, ^{137}Cs	Ryegrass	6 to 13	Kirchman et al. (1967)
Aqueous spray, ^{137}Cs	Wheat	14 to 31	Middleton (1959)
Fallout particles	Fescue	45	Dahlman (1971)
Fallout particles	Sorghum	38	Witherspoon and Taylor (1971)
Fallout particles	Fescue	69	Peters and Witherspoon (1972)
	Bluegrass	71	
	Bermuda	82	
	Zoysia	76	

Plant Uptake of Radiocesium from Soil

Soil serves as an effective sink for fallout ^{137}Cs, thus minimizing uptake by roots (see previous section on reaction of ^{137}Cs with clay minerals). Movement of radioactive cesium from soil to plants is dependent on the physical, mineral, chemical, and biological conditions of the soil. Other factors that govern uptake by plants are temperature, moisture, fertility, and pH conditions of soil. The soil–plant link limits the long-term movement of ^{137}Cs through the food chain to man. For example, after cropping for 520 days, less than 0.2% of the ^{137}Cs

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added to soil is transported to the foliar portion of legumes (Nishita, Steen, and Larson, 1958).

Concentration ratios, e.g., [^{137}Cs] plant/[^{137}Cs] soil, on a gram per gram basis, have been used to express the relative concentration of radionuclides in soil and plants. Radiocesium is considered to be slightly excluded by plants, namely, concentration ratios range from 0.01 to 1 (Menzel, 1965). Thus plants usually discriminate against rather than accumulate cesium from contaminated soil. The range of ratios reported for different plants growing in soil is given in Table 4. Radiocesium in plants grown on productive agricultural soils is represented by concentration ratios <1 (Nishita, Steen, and Larson, 1958; Nishita, Haug, and Hamilton, 1968; Evans and Dekker, 1968; Auerbach et al., 1959, 1972). However, from highly weathered acid soils, the concentration ratio of radiocesium is often >1 (Table 4, Evans and Dekker, 1969; Fredriksson, 1970b; Marei et al., 1972). For example, radiocesium uptake from acid podzol soils in the USSR and Canada is equal to the uptake of radiostrontium. The radiocesium concentration ratio between plants and the sandy acid soils of the southeastern United States and the lateritic Brazilian soils may exceed 10 in the absence of 2 : 1 layer silicates (Cummings et al., 1969; Fredriksson, 1970b; Sharitz et al., 1975).

Uptake of radiocesium from relatively unweathered productive agricultural soils is usually low (<0.5%). In natural environments uptake of ^{137}Cs from highly weathered soils may be equal to that of radiostrontium. Although relative concentration ratios range over at least 3 orders of magnitude, the higher values usually can be explained on the basis of low clay content, low pH, and a scarcity of clays possessing the 2 : 1 layer silicates. Soil fertility, abundance of organic matter, and management practices may also influence plant uptake of ^{137}Cs to a lesser extent, but an evaluation of these factors is beyond the scope of this discussion.

Radiocesium in Plants from Foliar Deposition vs. Root Uptake

It is important to recognize the relative contribution of ^{137}Cs in plants from direct foliar deposition vs. root uptake. Few data are available which permit a comparison of root vs. foliar uptake, but, from measurements of ^{137}Cs fallout and stable cesium ratios, Krieger, Kahn, and Cummings (1967) estimated a plant/soil ratio of 8×10^{-5} (discrimination factor) based on root uptake only (Table 5). Thus it would appear that in the absence of direct deposition the discrimination factor for ^{137}Cs in plants related to root uptake would be several orders of magnitude less than values given in Table 4. Russell and Bruce (1969) estimated independently that not more than 30% of the radiocesium burden in plants would originate from soil, and the real value might be closer to 10%. For example, only 3% of the ^{137}Cs burden in bluegrass is contributed by the root pathway in the Ohio region (Krieger, Kahn, and Cummings, 1967), and in

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TABLE 4

RELATIVE CONCENTRATION OF CESIUM IN PLANTS AND SOIL

Plant	Soil characteristics	Ratio*	Reference
Beans	pH 6.8; sand, 33%; silt, 46%; clay, 20%;	0.013 to 0.027	Nishita, Haug, and Hamilton (1968)
Red clover	178 Swedish soils	0.10 to 1.32	Fredriksson (1970a)
Oats	4 Canadian soils; pH 5.2 to 7.8; under different potassium and NH_4 fertility regimes	0.05 to 5.68	Evans and Dekker (1969)
Cereal, forage, and vegetable crops	pH 7.5; organic matter, 6.9%; cation-exchange complex, 24.2 meq/100 g	0.004 to 0.471	Evans and Dekker (1968)
Persicaria	Flooded and nonflooded conditions	0.003 to 1.35	Pendleton and Uhler (1960)
Grasses	Podzolic sand and podzol carbonate clays	0.3 to 8.2†	Marei et al. (1972)
Clover	Cropping 60 to 520 days; pH 6.8; sand, 33%; silt, 46%; clay, 20%	0.06 to 0.352	Nishita, Steen, and Larson (1958)
Winter rape	Brazilian laterite soil, pH 4.6; organic matter, 4.8%	0.89 to 13.2	Fredriksson (1970b)
Red maple leaves	Flood-plain sediment; predominantly kaolinite clay; sandy texture‡	0.2 to 5.4	Ragsdale and Shure (1973)
Sagittaria latifolia leaves	Flood-plain sediment; kaolinite-type clay, sandy texture‡		Sharitz et al. (1975)
	Low-level ^{137}Cs	20§	
	High-level ^{137}Cs	1.5	
Oats	Soils from southeastern United States	0.02 to 72¶	Cummings et al. (1969)
Millet	White Oak Lake bed soil (field plots)	0.03	Auerbach et al. (1959)
Millet	White Oak Lake bed soil (continuous cropping in pots)	0.03	Auerbach et al. (1972)
Aspergillus	White Oak Lake bed soil	0.01	Auerbach et al. (1974)

*Ratio of quantity in plant to quantity in soil on a per unit weight basis.

†Ratios based on units of pCi/liter in grass per pCi/kg in soil; thus, to compare with other ratios in Table 4, Marei's ratios should be adjusted upward to account for the possibility of a grass density of <1.0 kg/liter.

‡Data on soil texture and clay minerology taken from Brisbin et al. (1974), assuming that their data (Table 3, p. 24) from transects A and B would represent soil and sediment conditions for the delta (Sharitz et al., 1975) and the swamp forest (Ragsdale and Shure, 1973).

§The large ratio (20) may possibly reflect natural surface contamination of *S. latifolia* leaves by contaminated water or sediment because the species is typically emersed or submersed in aquatic environments.

¶Assuming at least 200 g of soil in 1-pt containers.

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TABLE 5

RELATIVE CONCENTRATION IN BLUEGRASS AND SOIL MINUS CONTRIBUTION BY DIRECT FOLIAR CONTAMINATION*

Basis	^{137}Cs (pCi) or stable Cs (mg/m^2) concentration		Ratio bluegrass/soil
	Soil	Bluegrass	
^{137}Cs-contaminated soil	79,000	8	0.00009
Stable cesium	750	0.055	0.00007

*Data from Krieger, Kahn, and Cummings (1967).

Colorado negligible ^{137}Cs (<1%) is taken up by alfalfa and corn from soil (Johnson, Wilson, and Lindsay, 1966).

Internal Dynamics of Radiocesium in Plants

Radiocesium is highly mobile once it gains entry to internal tissues of the plant (Resnik, Lunt, and Wallace, 1969; Middleton, 1959). Few data are available on the internal dynamics of radiocesium in plants with the exception of data from Middleton (1959), Levi (1969), Witherspoon (1964), Waller and Olson (1967), Brown (1964), and Dahlman, Auerbach, and Dunaway (1969). Data from Dahlman, Auerbach, and Dunaway (1969) illustrate the internal redistribution of an external tag of ^{134}Cs to fescue grass (*Festuca arundinaceae*) (Fig. 2). Extensive mobility is exhibited: 25% of the radiocesium absorbed by foliage is redistributed to roots over the initial 2-week period; approximately 25% moved to the inflorescence; at the time of flowering ^{134}Cs moved out of the roots into the inflorescence.

The first experimental data on the cycling of cesium in forest ecosystems were obtained from Witherspoon's (1964) experiments at Oak Ridge in 1960. When ^{134}Cs was introduced into the bole of white oak trees, 39% moved immediately to the canopy and 61% remained in the bole or was transferred directly via roots to soil. At the end of the second growing season, 18% of the original inoculum was in litter and 19% in soil. Cesium-134 in the foliar component was rapidly recycled as leaf fall and rainout. Over 50% of the leaf burden was returned to woody tissues before leaf fall. This experiment was the first of its kind to demonstrate the extensive mobility of cesium in the biological components of ecosystems.

In a separate study on ^{137}Cs cycling in tulip poplar, Waller and Olson (1967) found similar percentages remaining in the bole (woody compartment) after the second growing season (Table 6). The fraction that cycled through the

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canopy to the forest floor was also similar for both systems. More attention was given to underground processes in the tulip poplar experiments, and it was determined that 30% of the initial tag moved through the roots and was fixed in the soil before the end of the first growing season. By analyzing the ^{137}Cs concentrations in the clay fraction at various soil depths, Francis and Tamura (1971) confirmed that radiocesium originated from roots. The concentration in clay at the 6-cm depth was twice that at the 2- and 4-cm depths. The only

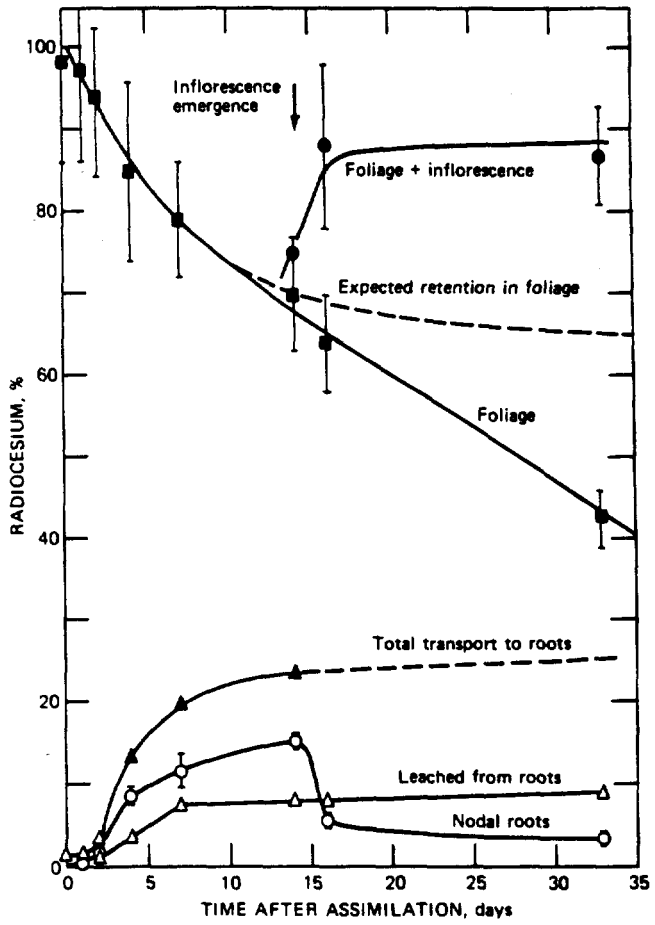

Fig. 2 Distribution of radiocesium in different organs of fescue following foliar assimilation. Cesium-134 was applied to foliage surface by emerging leaves in solution for 5 min. The extrapolated estimate of expected retention by foliage is based on an absence of transfer to developing inflorescence. The large error terms for foliage and inflorescence are attributed to variable initial retention on surface microsites and intercellular spaces.

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TABLE 6

DISTRIBUTION OF CESIUM IN PLANT AND SOIL COMPONENTS OF WHITE OAK AND TULIP POPLAR SPECIES AT THE END OF THE SECOND GROWING SEASON

Compartment	Percentage distribution*			
	White oak		Tulip poplar	
Bole	48	48	53	53
Canopy	(39)		(27)	
Roots	14		9	
			(30)	
Soil	18	51	36	47
Litter	19		2	
			(7)	
Export			2	2
Total		99		102

*Values in parentheses denote percentage of total cesium cycled through the compartment.

mechanism that explains the higher concentrations 6 cm beneath the surface is prompt transfer through the root pathway.

Distribution in root, soil, and litter components was different for the two systems (white oak vs. *Liriodendron*), but the summation percentages show somewhat similar concentrations in aboveground and belowground compartments (Table 6). The results illustrate the extensive initial mobility of cesium when it is introduced into trees. In both ecosystems, from 27 to 40% is cycled upward to the canopy, approximately 30% reaches the forest floor, approximately 30% moves directly to the root system, and most of that quantity moving to the roots appeared in soil in the tulip poplar community.

MODELS OF ENVIRONMENTAL TRANSPORT OF ^{137}Cs

Models of radiocesium dynamics in terrestrial ecosystems have been formulated using the extensive data bases on ^{137}Cs cycling in forests and on ^{137}Cs transport in the grass–cow–milk–man food chain. Another very meaningful application of models of ^{137}Cs dynamics is the prediction of the radiologic hazard to man using data on deposition, retention, and plant uptake from soil and concepts of food-chain transport and systems-analysis techniques. For example, Booth, Kaye, and Rohwer (1971) estimated radiocesium transport and the resultant dose to man for the soil–grass–cow–milk–man pathway of exposure. In their predictions concepts and data similar to those described in this paper were coupled with systems-analysis modeling techniques; ^{137}Cs

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concentrations in ecosystem components and dose to man were estimated for constant fallout input to the landscape, for equilibrium conditions, and for an average geographical location. The model estimates were compared with monitoring data for milk; agreement was relatively good because, for example, the predicted radiocesium content of milk was 62% of a 6-month observed average in 1970 (Booth, Kaye, and Rohwer, 1971). Such agreement would probably be acceptable for generic applications, and the accuracy of the model probably could be improved by considering site-specific parameters and by quantifying the nonlinear ecologic and climatic variations. In any event, it appears, on the basis of this successful application, that the concepts, techniques, data, and models do indeed describe the important relationships in terrestrial ecosystems. The quantitative data are adequate for modeling and describing the dynamics of ^{137}Cs in pathways leading to man.

RADIOCESIUM DYNAMICS IN ECOSYSTEMS OF THE SOUTHEASTERN UNITED STATES

The elevated cesium concentration in milk of the Tampa, Fla., milkshed has received close attention in the past decade (Pasteurized Milk Network, 1963–1973; Porter et al., 1967). Important questions related to this phenomenon are (1) Is sufficient information available to explain the reason for elevated ^{137}Cs in milk at Tampa? and (2) is a special mechanism responsible for increased uptake by vegetation (with a consequent increase in milk) such as that postulated by Gamble (1971)? Gamble contends that increased uptake is due to root–mycorhizal relationships. An assessment of these questions, using extant information on the biospheric behavior of ^{137}Cs, is required before additional research is commissioned. We submit that existing concepts and data are adequate to explain the elevated ^{137}Cs of the Florida milkshed. An assessment of the problem is made with information developed in previous sections of this paper.

The existing evidence indicates that direct deposition of fallout ^{137}Cs contributes substantially to the entry of ^{137}Cs into food chains. Wilson, Ward, and Johnson (1969) concluded that the concentration of ^{137}Cs in milk of the Tampa milkshed depended on both direct input from the atmosphere and on uptake from soil. It is interesting that their predicted values in milk decreased in relation to a lower air concentration of ^{137}Cs, and that the predicted and observed ^{137}Cs values for milk showed good agreement in the mid-1960s. That the close relationship of ^{137}Cs concentration in air and milk continued through 1973 (Fig. 1) is strong circumstantial evidence that direct deposition on vegetation still plays an important role in ^{137}Cs dynamics in the grass–cow–milk pathway.

Elevated levels of ^{137}Cs in Tampa milk are attributed to feeding dairy cattle Pangola (*Digitaria decumbens*) hay that contained 2 to as much as 10 times more

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^{137}Cs than other dairy-feed components (Porter et al., 1967). The mechanism of how Pangola hay accumulated more ^{137}Cs than other feeds was not determined, but both direct deposition on foliage and uptake by roots are the likely pathways, although substantial uptake by graminaceous plants may also occur at the base of tillers and stoloniferous structures on the soil surface (Russell, 1963).

For plants simultaneously exposed to both atmospheric and soil sources of ^{137}Cs, uptake via the root pathway would be expected to account for 1 to 3% (a maximum of 30%) of the plant burden. Cesium-137 uptake through plant roots is inversely related to the fixing capacity and clay content of soil and directly related to the sand content of 43 soils from the southeast, including at least five soils from Florida (Cummings et al., 1969, 1971). From their experiments, greatest plant uptake is observed for humic gley-type soils (characterized by highly weathered kaolinitic clays and sandy texture). An average of 2.8% of added ^{137}Cs was assimilated into oat seedlings in pot culture. These observations are consistent with extensive literature data (Table 4), namely, higher ^{137}Cs uptake (ratios >1) is reported for sandy, acid, highly weathered soil, particularly in the absence of 2 : 1 expanding lattice clays.

The evidence indicates that direct deposition of ^{137}Cs on foliage and appreciable root uptake (2.8% of ^{137}Cs in soil) may both be responsible for increased levels of ^{137}Cs in Florida milk. It is difficult to delineate the exact responsible mechanisms because no deliberate measurements have been made of direct deposition of fallout on Florida vegetation, its retention on foliar surfaces, and the subsequent incorporation into livestock feed. In continental environments direct deposition appears to account for at least 95% of the ^{137}Cs content of forage (Johnson, Wilson, and Lindsay, 1966; Kreiger, Kahn, and Cummings, 1967). Ward, Johnson, and Wilson (1966) estimate a net deposition velocity (V)* of 0.001 m/sec, and their results indicate that at least an equivalent quantity of ^{137}Cs is deposited by rainfall. According to Gifford and Pack (1962), a value of 0.001 m/sec for V probably is realistic for ^{137}Cs.

Upper and lower limits of ^{137}Cs deposition are estimated in Table 7. When we recognize that direct deposition is affected by ecosystem features (vegetation surfaces and density, soil exposure) and meteorologic conditions (rainfall, atmo-

*Net deposition represents the fraction on vegetation after weathering losses have occurred. It represents the quantity available for subsequent ingestion by a consuming animal. It is a convenient expression because its determination does not require knowledge of fractional retention, kinetics of weathering, and intraplant redistribution. Further, net deposition velocity (V) is conveniently used with air concentration to estimate ground concentration (D), i.e., $D = VAt$, where A = air concentration (curies per cubic meter) and t is time in seconds. The units of V are meters per second and of D are curies per square meter.

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TABLE 7

ESTIMATED RELATIVE CONTRIBUTION OF ^{137}Cs TO VEGETATION VIA ATMOSPHERIC DEPOSITION AND ROOT UPTAKE

Duration, days	Source	^{137}Cs concentration, pCi/m^3 air or pCi/kg soil	^{137}Cs concentration in vegetation: Direct deposition, pCi/kg: Dry*	Total†	Root uptake,‡ pCi/kg
30-day growing season	Air§	5.6×10^{-3}	56	112	
300	Air	5.6×10^{-3}	560	1120	
Growing season	Soil¶	205			860

*Dry deposition (D) is calculated according to $D = A \times V \times t/M$, where A = air concentration (see footnote §), V = deposition velocity = 0.001 m/sec, t = time in seconds (2.6×10^6 sec per 30 days and 2.6×10^7 sec per 300 days), and M = mass of vegetation in kg/m^2 (0.2 kg/m^2 for unfertilized pasture).

†Total deposition (dry + wet) is approximately twice dry deposition according to Ward et al. (1966).

‡Root uptake (R) is calculated from $R = S \times f \times Z$, where S = soil concentration (see footnote ¶), f = fraction of total assimilated [2.8% according to Cummings et al. (1969, 1971)], Z = mass of soil in effective rooting zone, a 10-cm depth was selected for this analysis. A diffuse root system extends deeper than 10 cm, but Gamble (1971) shows most of the ^{137}Cs in the top 10 cm of soil.

§Air concentration in pCi/m^3; average of midyear values for 1969, 1970 data (Fig. 2) and HASL (1974), pp. B-78, B-79.

¶Soil concentration taken from Gamble (1971).

spheric mixing), we realize that the estimates are by no means exact; they should be regarded as approximations for purposes of discussion. These calculations suggest that we would expect only a nominal direct deposit of radiocesium on foliage over a 1-month growing period, but for longer periods, e.g., 10 months, dry deposition could account for possibly more ^{137}Cs in vegetation (1120 pCi/kg) than would be expected from root uptake (860 pCi/kg), using the average percentage uptake (f) of 2.8% (Cummings et al., 1969). This percentage uptake value is an order of magnitude greater than the upper-limit percentage uptake commonly reported for agricultural soils (0.3%; United Nations Scientific Committee on the Effects of Atomic Radiation, 1962). Thus, for either a lower f value or for a higher V, the direct-deposition mechanism would account for

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substantially more ^{137}Cs in forage than that attributed to root uptake especially when chronic fallout deposition is estimated over a full growing season or longer.

In Table 7 total ^{137}Cs in forage is estimated for both direct deposition and uptake from soil, and the estimates are compared with measured values reported for southern Georgia and Florida. Total estimated ^{137}Cs in Florida plants, approximately 2000 pCi/kg for both direct deposition and root uptake, compares favorably with many reported values for vegetation growing on humic gley-type soils in southeastern Georgia (Cummings et al., 1971). The average ^{137}Cs concentrations reported for leaves were 1000, 1800, 1600, and 2800 pCi/kg for gallberry, wax myrtle, smilax, and swampberry, respectively. The grand average (1800 pCi/kg) for these species is in good agreement with estimated deposition and uptake for a growing season (2000 pCi/kg). The average ^{137}Cs concentration for mixed grasses was 6100 pCi/kg, three times the estimated 2000 pCi/kg. The discrepancy between estimated and observed concentrations may be due to a higher effective deposition on grass foliage or possibly to increased uptake at the base of graminaceous plants, as described by Russell (1963). Substantially higher values (factors of 5 to 10) were observed for leaves of red bay, palmetto, holly, and deer's tongue and for mosses. The discrepancies between observed and estimated values for these species are not easily explained, although local soil factors (acid pH, low clay content, absence of 2 : 1 expanding lattice clays) or long periods (several growing seasons) of direct deposition on evergreen foliage may account for some of the difference. Total estimated ^{137}Cs in vegetation (2000 pCi/kg) also agrees favorably with a range of values that Gamble (1971) reports for Florida vegetation in 1969 and 1970: for pine needles, 270 pCi/kg; scrub oak leaves, 2700 pCi/kg; palmetto fronds, 2400 pCi/kg; hardwood forest leaf litter, 900 and 1300 pCi/kg, for a grand average of 1500 pCi/kg.

It appears from this assessment that the elevated concentrations of ^{137}Cs in vegetation of coastal-plain ecosystems and for the Tampa, Fla., milkshed can be explained on the basis of current knowledge of radiocesium dynamics in terrestrial environments. The combination of direct deposition of ^{137}Cs on vegetation and the high fractional transfer (approximately 3%) in the soil-root pathway can account for the levels of ^{137}Cs observed in many species that grow on sandy, acid, kaolinitic soils—sometimes classified as humic gley's—of Florida. Low cesium-fixing capacity due to mineralogical characteristics and low clay content are probably important soil factors responsible for increased plant uptake. A new, yet unidentified factor, e.g., mycorhizal, symbiotic enhancement of uptake (Gamble, 1971), is not required to explain the elevated levels of ^{137}Cs in vegetation and milk, although the possible existence of such a mechanism is not disputed. Direct deposition of fallout on vegetation appears to be an important mode of contamination; otherwise the concentration of ^{137}Cs in milk—a function of concentration in forage—would not decrease in tandem with the order of magnitude decrease in ^{137}Cs in the atmosphere which has been reported in the past decade.

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