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RADIATION RESEARCH 94, 135–150 (1983)

Radionuclide Transport Processes in Terrestrial Ecosystems[1]

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WHICKER, F. W. Radionuclide Transport Processes in Terrestrial Ecosystems. *Radiat. Res.* **94**, 135–150 (1983).

Some major principles and the status of knowledge concerning the transport of radionuclides through terrestrial ecosystems are reviewed. Fundamental processes which control the flow of radionuclides between ecosystem components such as air, soil, plants, and animals are described, with emphasis on deposition, resuspension, plant uptake, ingestion, and assimilation. Properties of radionuclides, organisms, and ecosystems are examined in relation to their influence on the accumulation of radioactive materials by plants and animals. The effects of the physicochemical nature of the radionuclide; morphology, physiology, and behavior of the organism; and soil, nutrient, and trophic characteristics of the ecosystem are highlighted. Observations in natural ecosystems on radionuclides such as ^{137}Cs, ^{90}Sr, ^{131}I, ^{3}H, and ^{239}Pu are used to illustrate current concepts. An assessment of the degree to which the processes controlling radionuclide behavior are understood and of our ability to simulate and predict such behavior with computerized models is offered. Finally, brief comments are made on research needs.

INTRODUCTION

Radiobiological effects in organisms which result from environmental radioactivity are dependent on the radionuclide concentrations in critical biological tissues and adjacent media. The degree of concentration is determined by ecological and physiological transport processes, and properties of the radionuclides involved. The purpose of this paper is to review some of the major processes and principles which control the behavior of radionuclides in the terrestrial environment. These processes are natural phenomena which accomplish fundamental transfers of energy and nutrients in ecosystems. They may be physical, chemical, or biological in character, and they produce highly nonuniform, but generally predictable concentrations of radionuclides in organic and inorganic material.

The detailed understanding and ultimate prediction of temporal distributions of radionuclides in the environment are difficult to achieve because of the complexity of ecosystems; the behavioral, morphological, and physiological intricacies of organisms; and the chemical and physical dissimilarities among the many radionuclides of concern. Despite such complexity, there has been sufficient radioecological progress over the last 25 years to enable the formulation of numerous generalizations and the development of reasonably mechanistic and accurate simulation models. However,

[1] Presented at the Symposium on Transport of Radionuclides in Our Terrestrial Environment at the 29th Annual Meeting of the Radiation Research Society, Salt Lake City, Utah, April 19, 1982.

0033-7587/83 $3.00
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as a result of water infiltration, mechanical sifting, and biological activities. Surface deposits also tend to be covered by the accumulation of organic litter and aerosol deposits. Such phenomena reduce the potential of resuspension but increase the opportunity for uptake by plant roots. Root uptake of radonuclides from soil is a highly complex process which is subject to many variables (*16, 17*). Generally, the process is significant mainly for longer-lived radionuclides which exist in soluble chemical forms and do not become tightly bound by the soil. In addition, soil water and metabolic activity by the plant are necessary for active root uptake. The propensity for a radionuclide to be taken up by a plant from the soil is usually expressed by a "concentration factor." This parameter, defined as the concentration in the plant divided by the concentration in the soil, is dependent on radionuclide, chemical form, plant species, soil properties, and numerous other factors. If radioactive material migrates below the root zone or becomes so tightly bound by soil particles that root uptake is precluded (*18*), it is said to have entered a "sink" compartment.

Terrestrial plants usually accumulate radioactivity by two principal mechanisms, namely, direct deposition from the atmosphere and root uptake (*19, 20*). Direct deposition generally dominates the two processes, especially soon after a contamination event. When terrestrial plants are in contact with standing water, direct sorption of dissolved or suspended radioactivity to vegetative surfaces can also be important. The differences in contamination mechanisms often lead to the need for two aboveground plant compartments in simulation models. The plant interior may be treated as the compartment which receives materials through internal translocation following root uptake, whereas the plant surface receives contaminants from aerial deposition or surface sorption. Some surficial radioactivity may be absorbed into the plant interior, depending upon the radionuclide and its chemical form. Some radioactivity of foliage is released back to the environment, the mechanism again dependent upon which plant compartment is considered. Surficial deposits weather from foliage, largely through mechanical agitation from wind or other disturbances, or through the washing or leaching effects of rain (*20*). Both internally and externally incorporated radioactivity drop to the ground in the form of organic detritus from leaf-fall.

The usual principal route of entry of environmental radioactivity into herbivorous animals is the ingestion of contaminated foliage (*20–22*). The rate of intake is proportional to the feeding rate and to the concentration of radionuclide in the forage. Since different plant species vary considerably in their tendency to accumulate radioactivity, the feeding habits and preferences of herbivores play an important role in determining the quantity of radionuclide ingested. The fraction of the ingested radioactivity which is assimilated and transported to various tissues and organs is dependent upon the physical and chemical properties of the radionuclide, properties of the ingesta such as abundance of competing elements, and the physiology of the animal (*22, 23*). In some cases, assimilation of a radionuclide incorporated within the plant interior is greater than that of the same radionuclide on the plant surface, presumably owing to differences in physicochemical form (*24*).

Competing with ingestion of forage as a route of radionuclide entry to herbivores are ingestion of soil and inhalation. In most natural situations, ingestion of plant material is much more important than inhalation as a route of entry to herbivores

(*20, 21*). However, inhalation should not be dismissed as a source until it can be rigorously demonstrated that such is warranted, for certain circumstances could result in exceptions to the rule. Many herbivores ingest significant quantities of soil, sometimes inadvertently and sometimes purposefully (*25–27*). Consumption of dusty plant matter, grooming, and grazing close to the soil surface can lead to inadvertent soil ingestion. Soil may also be ingested directly, possibly as a source of elements. Even though the mass of soil consumed is usually less than 10% of the dry forage mass ingested, soil intake can be relatively important if most of the ecosystem inventory is on the ground surface. Typically, soil ultimately becomes the major reservoir of longer-lived radionuclides in terrestrial ecosystems (*28*).

Excretions, secretions, molting, and mortality all contribute to the transfer of radioactivity from animals to the detritus and soil compartments. The concentrations of radionuclides in animal tissues are as strongly influenced by the rates of excretion as they are by intake and assimilation. The rate of excretion is affected by a host of variables, including the radionuclide and its chemical form, site(s) of deposition, intake rate of competing elements, and physiological factors (*23*). Animal tissue concentrations are roughly proportional to the rate of intake and the assimilation fraction, and inversely proportional to the rate of excretion.

The processes governing radionuclide accumulation in carnivores and omnivores are basically the same as those for herbivores. The major differences result largely from food habit departures. The situation is more complex for humans and certain other animals that subject food to washing and preparation. In general, washing and preparation somewhat reduce the concentrations of most radionuclides in foods (*29*). Food storage is common not only in human agricultural food chains, but also in a number of natural food chains. Where it occurs, storage provides opportunity for physical decay of the shorter-lived radionuclides.

The compartments and processes just described in qualitative language and illustrated in Fig. 1 can be portrayed mathematically (*30*). Each process may be formulated as a rate of radionuclide flow (R) in units of activity per unit time per unit area. The time derivative of the radionuclide inventory in each of h compartments (I'_h) may be stated as a differential equation,

$$I'_h = \sum_{i=1}^{n} (R_{\text{in}})_i - \sum_{j=1}^{m} (R_{\text{out}})_j,$$

where $(R_{\text{in}})_i$ denotes the ith rate of inflow to compartment h and $(R_{\text{out}})_j$ denotes the jth rate of outflow from compartment h. A differential equation can thus be written for each compartment. Solution of the resulting set of differential equations (usually by numerical algorithm) yields time-dependent radionuclide inventories (activity per unit area) for each compartment. Inventories may be converted to concentrations, which in turn can be used for dose assessment, or for comparison to real data as a test of the model's simulation ability.

FACTORS AFFECTING RADIONUCLIDE ACCUMULATION

Observations over the past two or three decades have provided sufficient data to encourage the proposition of numerous generalities concerning the behavior of ra-

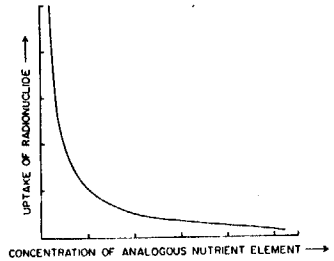

FIG. 3. The biological uptake of radionuclides which are chemically similar to abundant nutrient elements tends to be inversely proportional to the available concentrations of corresponding nutrients in the environment, thus giving rise to a hyperbolic relationship.

elements of interest in Fig. 4. The reader is urged not to use these data for serious calculations because the ranges of values are very large and dependent on numerous factors, some of which are discussed in this paper.

Organism Characteristics

Innate attributes of plants and animals can markedly affect the degree to which they accumulate radionuclides. Concentrations in different species within the same community can vary significantly, despite the fact that they may be exposed to a common source of contamination. The explanation of such variations can usually be based on differences in morphology, longevity, behavior, and physiology.

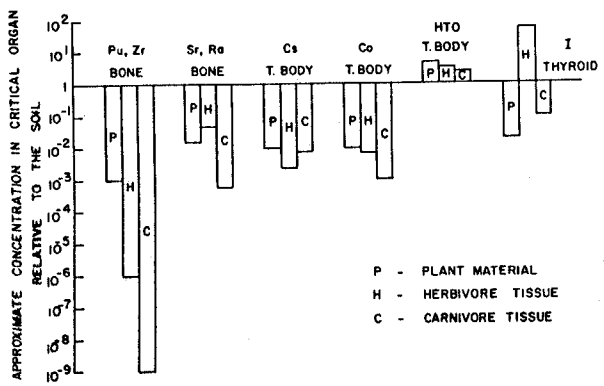

FIG. 4. Approximate concentrations of selected elements expected in critical biological tissues relative to a unit concentration in soil. Substantial departures from the values given occur in specific cases.

External morphology is frequently a major determinant of the tendency to accumulate radioactivity (*14*). When surface adsorption is a major vector, organisms which present a large surface-to-volume ratio tend to contain more radioactivity per unit mass than biota with a small ratio. This means that especially small organisms and those with surfaces which are highly folded or contain hair or other projections efficiently accumulate environmental radioactivity on their surfaces. Small arthropods and thin or pubescent foliage are good examples which illustrate this phenomenon. Lichens provide another excellent example in that their highly flattened, convoluted, or branched forms, adapted for adsorption and subsequent absorption of nutrients from the atmosphere, present a large surface-to-volume ratio. As a result, lichens adsorb radionuclides from air and precipitation with remarkable efficiency.

Another feature of lichens which is perhaps equal in importance to morphology is longevity. Being extremely slow growing and long-lived, lichens tend to retain nutrients and radionuclides tenaciously. Because of this, the lichen tends to accumulate long-lived radionuclides over periods of years or even decades, and very high concentrations can ultimately result from comparatively low level, but chronic, exposure (*36*). Bone of long-lived vertebrates provides another example of a tissue which will accumulate and efficiently retain certain radionuclides over periods of many years. Such long-term buildup is mainly relevant to longer-lived, bone-seeking nuclides which are continually ingested over a significant length of time. The annual leaf-fall characteristic of deciduous trees and shrubs precludes these otherwise generally efficient accumulators of radioactivity from achieving the exceptionally high concentrations found in lichen and other persistent tissues. An example of the differences between various species of vegetation in the ability to accumulate radioactivity is provided by the author's data on fallout ^{137}Cs gathered in 1964 in the north-central mountains of Colorado (Fig. 5).

Behavioral factors of relevance to radionuclide accumulation in animals include migratory movements, habitat selection, food preferences, and activities such as grooming, burrowing, etc. A dramatic example of the combined effects of migration, habitat use, and food selection is the seasonal accumulation of ^{137}Cs in Alaskan caribou (*37*). During the fall migration, caribou move southward to areas where winds sweep the landscape relatively free of snow and lichens are abundant. The remarkable ability of lichens to accumulate ^{137}Cs, owing to morphology and longevity as mentioned earlier, and also their comparatively low potassium content, leads to abrupt, dramatic increases of the radionuclide in caribou flesh (Fig. 6). In spring when the caribou move northward, dietary changes from lichens to higher plants reduce the intake of ^{137}Cs substantially and the tissue burdens plummet.

Another example of the influence of food habits on the accumulation of environmental radioactivity by animals was provided by Osburn (*14*) who studied numerous bird and mammal species within a single watershed of Colorado's Front Range from 1962–1964 (Fig. 7). The measurements of gross β radioactivity were made on the skeleton + musculature and primarily reflected fallout radionuclides such as ^{137}Cs and ^{90}Sr. It is clear that carcasses of predatory and seed-eating species were comparatively low in radioactivity, while grazers and insectivores exhibited elevated tissue concentrations.

Metabolism and other physiological functions also play a fundamental role in the

extensively taken up by vegetation. Radiocesium follows the same pattern. It is readily available to the plant because it does not become fixed by the soil, and the high water, low potassium levels enhance root absorption. Thus plants of the region are comparatively high in ^{137}Cs and low in potassium. This situation is likewise ideal for maximizing accumulation of radiocesium in animals. The fact that ^{137}Cs is long-lived and efficiently recycled into the biota explains the observed temporal persistance of the nuclide in plants and animals of the region.

A comparison of ^{137}Cs concentrations in muscle tissues of North American beef, deer, and caribou during the period 1968–1970 is given in Fig. 8. The values observed by Hanson (*37*) for caribou were multiplied for 3.5 to account for the three- to fourfold lower worldwide fallout deposition in the arctic as compared to the continental United States. It is clear from this contrast that properties of ecosystems produce dramatic differences in biotic concentrations of radionuclides from similar levels of deposition.

The structure of biotic communities is affected in part by features such as vegetative biomass, species diversity, and trophic relationships. Such characteristics can exert considerable influence on radionuclide behavior. For instance, communities having characteristically low vegetative biomass such as arctic and alpine systems tend to exhibit relatively high radionuclide concentrations simply because the radionuclide inventory is contained within a small mass. The typically prostrate, low-growing plants of these regions are also in intimate contact with melting snow which enhances direct transfer of radionuclides to the vegetation. Conversely, densely vegetated areas such as forests tend to distribute radioactivity over a much larger mass, leading to reduced concentrations.

Ecosystems with a high degree of species diversity tend to exhibit relatively complex food web relationships. The flow of minerals, energy, and radionuclides through such systems will be highly partitioned and apportioned among many species. Ecosystems of low diversity tend to channel a greater fraction of the radionuclide flow through a few species which are functionally dominant.

The majority of radionuclides, owing to their chemical and physiological properties,

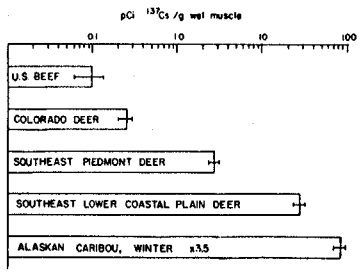

FIG. 8. Mean (±1 SE) ^{137}Cs concentrations in beef cattle, deer, and caribou from various ecosystems of North America during 1968–1970. The values for Alaskan caribou were multiplied by 3.5 to account for lower fallout deposition in the arctic as compared to the continental United States. Data are from various sources (*37, 39*, author's).

tend to decrease in concentration with trophic level (Fig. 4). In other words, the relative concentrations generally follow the scheme: plants > herbivores > omnivores > carnivores. A well-known exception to this rule is ^{137}Cs, which has been shown to generally increase with trophic level, particularly when the relative nuclide levels are expressed as the $^{137}Cs/K$ ratio in the higher trophic level divided by the $^{137}Cs/K$ ratio in the lower trophic level. As a rule, the trophic ratio (the ratio of the two $^{137}Cs/K$ ratios) achieves a value of roughly 2 or 3 (*41*). The fundamental reason for this appears to be the more tenacious retention of cesium in tissues as compared to potassium. Whereas Cs and K generally have comparable assimilation fractions, the biological half-time for Cs is two or three times as long as that for K. Another prominent exception is the concentration of radioiodine in the thyroid gland of herbivores (Fig. 4).

STATUS OF PRESENT KNOWLEDGE

Research in areas related to radionuclide transport in terrestrial ecosystems began with small, isolated efforts in the early 1940s. During the late 1940s and 1950s, development of nuclear weapons programs caused dramatic growth in various aspects of radioecology. Such growth was sustained in the 1960s, largely as a result of progress in the nuclear power industry. In the 1970s, despite curtailments in growth of the nuclear power industry in the United States and diminished levels of nuclear weapons fallout, viable programs in the field were generally maintained. This probably owed in large measure to increased environmental awareness and regulation, growing medical and other applications of radioiosotopes, and persisting concerns over nuclear issues such as weapons and waste disposal.

As of the present, I would guess that there are of the order of 10^4 open literature publications specifically relevant to radionuclide behavior in terrestrial ecosystems. However, there have been relatively few concerted attempts to synthesize this diverse and widely scattered material. One notable exception is in the area of transport of transuranium radionuclides (*42*). Other exceptions include general books on radioecology by Russian (*43*) and American (*44*) workers. The latter reference in particular provides a much more comprehensive guide to the literature on radionuclide transport in terrestrial ecosystems than does this review.

The depth of understanding of radionuclide transport processes is considerable, but the magnitude and complexity of the field leave much room for future progress. For example, certain workers and laboratory groups are intimately familar with the behavior of a very limited number of radionuclides in but a very few ecosystems. Such familiarity has often been extended to the construction of mechanistic transport simulation models (*45, 46*). The validation and testing of such models provide some confidence that our understanding, though not complete, is substantial. On the other hand, radionuclide behavior in certain types of ecosystems (e.g., alpine tundra, montane forests, shortgrass plains) has scarcely been investigated and some nuclides (e.g., natural uranium and progeny) have received but minimal attention (*46*).

The answer to the question of research needs is tempered by operational priorities and transport parameter sensitivity analyses, in addition to the status of pure knowledge. The pattern of the past three decades has seemed to have been to support research first on the basis of perceived problems or needs, and secondarily on the

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RADIATION RESEARCH 94, 151–155 (1983)

The Distribution of Tritium among the Amino Acids of Proteins Obtained from Mice Exposed to Tritiated Water[1]

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COMMERFORD, S. L., CARSTEN, A. L., AND CRONKITE, E. P. The Distribution of Tritium among the Amino Acids of Proteins Obtained from Mice Exposed to Tritiated Water. *Radiat. Res.* **94**, 151–155 (1983).

The distribution of tritium among the amino acids of serum proteins in mice chronically exposed to tritiated water was determined by ion exchange chromatography of the protein hydrolysate. The specific activity of nonexchangeable tritium in these amino acids relative to the specific activity of tritium in the tissue water of mice ranged from 0.04 for phenylalanine and threonine to 1.0 for glycine and alanine. Since tritium from tissue water can enter the nonexchangeable positions of amino acids only as the result of metabolic processing, the relative specific activity of tritium in each amino acid is an indicator of the extent of such processing. The tritium content of tyrosine and all the amino acids required in the diet for survival is quite low, except for histidine, and can be entirely accounted for by transamination or, in the case of methionine, by transmethylation. The tritium content of the other amino acids is too high to result from such minor processing and must reflect primarily the fraction synthesized *de novo*. The implications of these findings with respect to the radiobiological consequences of a diet containing tritiated proteins are discussed.

INTRODUCTION

After exposure of animals to tritiated water, only the tritium incorporated into nonexchangeable positions in histones and DNA or present as water in the tissue fluids needs to be considered when calculating the dose to cell nuclei (*1, 2*). The amount of tritium in other cell components can be ignored because it is either too small, too remote from the cell nucleus, or turning over too rapidly.

When animals are exposed to tritium through their diet, rather than through their drinking water, the situation is expected to be more complex. An important factor in determining the degree of complexity is the extent to which the tritium in proteins ingested by an animal exchanges with hydrogen in tissue water rather than transferring directly to newly synthesized proteins. In the absence of isotope effects, the fraction of ^{3}H in amino acids which exchanges for ^{1}H in the tissue water of animals receiving tritiated amino acids and normal water should be equivalent to the fraction of ^{1}H in amino acids which exchanges with ^{3}H in the tissue water of animals receiving unlabeled protein and tritiated water. We have measured this by isolating serum

[1] Research supported by U. S. Environmental Protection Agency Agreement EPA-79-D-X0533 and U. S. Department of Energy Contract DE-AC-02-76CH00016.
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