CONF-8609191--2

CONF-8609191--2

DE87 001836

# COMPARATIVE BEHAVIOR OF THREE LONG-LIVED RADIONUCLIDES IN FOREST ECOSYSTEMS

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## Introduction

Contamination of large land areas by radionuclides has been limited to sites of nuclear facilities, bomb testing, or accidents such as that at Chernobyl and the one in the eastern Ural Mountains [1,2]. Fortunately, such situations have been limited in number. Likewise, our information about the long-term ecological consequences attendant upon such situations is limited. Environments containing curie quantities of long-lived radionuclides do provide an opportunity to learn more about the long-term distribution of these materials in a natural ecosystem. The importance of this question derives from many obvious concerns (health impacts, ecological impacts, land-use practices, and cleanup methods), but an overall question is: What shall be the policies for long-term management of contaminated land?

To arrive at answers to that complex question, we need an understanding of the approximate steady-state distribution of long-lived radionuclides in contaminated ecosystems, recognizing that we must be prepared for exceptions and differences arising from intrinsic differences in ecosystem properties. At Oak Ridge National Laboratory (ORNL) various operations associated with nuclear energy research and related waste disposal activities have resulted in releases of fission products and other radioisotopes to local environments, especially during the 1940s and 1950s. This local contamination served as a stimulus for carrying out many investigations on the consequences and long-term fate of radionuclides in the environment. Some local forest ecosystems were contaminated with isotopes of $^{239,\ 240}Pu$, $^{99}Tc$, and $^{137}Cs$ as well as others, many of which, being short lived, have not been the subjects of continuing investigations.

## Description of Contaminated Ecosystems

This paper deals with studies in three forest ecosystems in eastern Tennessee, an area of rich temperate deciduous forests, sometimes referred to as mixed mesophytic forests. Two of these forest ecosystems were contaminated as a result of waste disposal operations. The third was experimentally tagged with millicurie quantities of $^{137}Cs$.

One of these ecosystems is a floodplain forest that is typical of this region. This forest has been growing on alluvial soils since 1944. Prior to that time the area was a temporary holding pond within White Oak Creek which received radioactive effluents from ORNL. Radiocesium was

deposited in the pond sediments as were $^{90}Sr$, $^{239}Pu$, $^{241}Am$, and other radionuclides. The dam which created the pond failed in late 1944, and the area was allowed to revert to natural conditions. The result was the development of a floodplain forest consisting of three different forest communities [3,4]. The soils are fertile alluvials representative of bottomlands. The overstory tree species are principally ash, sycamore, boxelder, willow, and sweetgum (Fraxinus americana L., Plantanus occidentalis L., Acer negundo L., Salix nigra Marsh, and Liquidambar styraciflua L., respectively).

This floodplain forest ecosystem represents a situation in which the source of contamination is in the soils and sediments. An extensive data base on radiocesium concentrations, based on the intensive surveys and analyses carried out, is available [3,4]. The mean radiocesium concentrations in the soil range from a few hundred picocuries per gram to over 80 000 pCi per gram, depending on location. The plutonium inventory for the contaminated portion of this floodplain is approximately 0.6 Ci, with an average soil burden of $1.6 \times 10^7$ pCi/$m^2$ [5].

The situation at the second forest site, which was contaminated by $^{99}Tc$, is more complicated. The wooded area lies within a small ravine drained by a stream. The predominant tree species here are red maple (Acer rubrum L.), tulip-poplar (Liriodendron tulipifera L.), and elm (Ulmus americana L.). The soil is a silty clay loam with a pH of 5.5, which is typical of bottomland soils in this region of Tennessee. Approximately 90 m to the east of the forest is located an asphalt-covered underground burial trench, which was used in the past for disposal of intermediate-level liquid radioactive wastes. Beginning in 1960, this pit received more than 300 000 Ci of mixed fission product wastes, an unknown proportion of which was $^{99}Tc$, before it was covered and capped in 1966 [6]. An early tree monitoring survey carried out in 1962 showed that the trees west of the perimeter of the pit site were being contaminated, and further analysis demonstrated that there was groundwater seepage occurring in a westerly direction toward the ravine forest site [7]. Because of the shallow water table, it has been difficult to apportion the source of the biologically available $^{99}Tc$ between the groundwater and the soil, especially since the roots of the trees in this area may in part be in direct contact with the groundwater at certain times of the year. The concentrations of $^{99}Tc$ in

given ecosystem should not change significantly as a function of time. From Table I it is evident that even order-of-magnitude changes in $^{137}Cs$ concentrations in compartments other than soil will have a negligible effect on the overall total percentage in the soil.

Table II presents a comparison of the estimated steady-state distribution of technetium, plutonium, and cesium among the various compartments of a deciduous forest. The technetium and plutonium inventories come from previous studies [8,10]. The method used for calculating the distribution in the ecosystem was the same in each case. First, the grams (dry weight) of biomass per square meter of ground surface was calculated from regression equations which use tree diameter at breast height as an independent variable [11]. The diameter at breast height was measured for all trees in the technetium study area and in the floodplain forest. The biomass estimates were then multiplied by concentration data to obtain an estimate of radioactivity per square meter. Estimates of activity per unit area were added up for all compartments including soil. A standard soil depth of 20 cm was used for the floodplain forest and the technetium study area to facilitate comparisons. A soil density of 1.3 $g/cm^3$ was assumed. Assumptions pertaining to soil depth may have caused underestimation of the activity present in the soil because radiocesium (and probably plutonium) can be found deeper than 20 cm in the floodplain forest.

In analyzing these data, one must be careful to distinguish among those differences which are due to basic ecosystem properties, those which may be the result of different chemical species, and those which are due to differing sampling techniques and analyses. For example, there is a striking difference in the aboveground tree compartment (leaves and stems) for the different radionuclides, which reflects the differences in isotopic properties, as in the case of technetium (8.0%) and cesium (0.021%), although both forests are growing on similar types of soil. This difference contrasts with the radiocesium percentages (8.5 vs 0.021%) for the aboveground tree compartment of two different forests. Here the differences are related to the kinds and amounts of clay present in the soils. While this phenomenon is generally well known and is the basis for concerns about radiocesium fallout in areas with poor or sandy soils, such as in the Arctic regions, the data illustrate the importance of not generalizing; rather, we need to understand and be able to

quantify the cycles for those ecosystems which differ in their basic biogeochemical characteristics.

Differences between the litter compartments and the stump and root compartments reflect either differences in sampling methods, in analysis, or difficulties in measurement. Litter for plutonium analysis was collected from the ground surface, whereas the litter in the cesium study of the floodplain forest was collected by means of aboveground collectors, which precluded leaves from having any contact with the contaminated soil. Similarly, in the case of the root compartments, comparisons across ecosystems carried out by different investigators or by the same investigators at different times using varying techniques should be viewed with caution. Concentrations in roots as well as tree root biomass are difficult parameters to estimate. The differences between root values in Table II reflect these types of problems. Therefore, one should not attribute great significance to the differences between ecosystems when dealing with variables which are difficult to quantify accurately.

## Models

Systems models provide a convenient and accurate means for summarizing data on inputs and transfers of radionuclides between the structural and functional components of an ecosystem. While current interest in the use of models is directed toward their use as predictive tools, they are equally valuable for synthesis because they provide a common framework for analyzing changing radionuclide accumulations within different compartments of an ecosystem, thereby enabling evaluation of transfer coefficients between different radionuclides and between different ecosystems.

A number of models of radionuclide behavior in forest ecosystems have been developed for different radioisotopes [10,12,13]. These have all proved useful in enabling us to understand and predict the behavior of radionuclides in a forest ecosystem. Most recently Garten [14] has developed a six-compartment, linear, donor-controlled model which lends itself well to summarizing and comparing radionuclide data from the literature (Fig. 1). The model is designed to account for inputs to the forest ecosystem either directly through injection or indirectly by way of fallout or from subsurface sources such as buried wastes. For purposes of this paper, the soil is treated as a single compartment

because of limited data and because the question of what constitutes available and unavailable pools is heavily dependent on the particular analytical methods used, the nature of the soils, and other factors. It is still a question that can be understood in principle but varies from site to site. All other compartments are typical of other types of forest models. Transfer rates were derived using data from the literature.

Nine transfers were included in Garten's model (Table III). Average annual biomass values [g (dry weight)/$m^2$] and radionuclide concentrations [pCi/g dry weight)] were multiplied to calculate the inventory of radionuclides (pCi/$m^2$). Transfer coefficients in the model were calculated on the basis of biomass flux ($g \cdot m^{-2} \cdot year^{-1}$) from the donor compartment. Where specific data were lacking for these forest sites, fluxes were derived from biomass data collected in the Oak Ridge area during the International Biological Program [11]. A few parameters were arrived at through parameter fitting during simulations with the model forests. The forests from which the data for the model and for these comparisons were obtained were the aforementioned floodplain forest ecosystem, the forest ecosystem contaminated by $^{99}Tc$, and the tagged tulip-poplar (Liriodendron) forest. The tagged tulip-poplar forest represents a special case that is somewhat atypical for comparative purposes because the radioisotope was introduced directly into the tree. Such direct tagging made possible rapid and accurate measurements of intra- and interseasonal transfers of radiocesium, thereby providing early insight into the rates of transfer within the biotic components of the ecosystem. By the end of the first growing season and during the subsequent several years of sampling, it became evident that the bulk of the radiocesium was being cycled to and retained by the soil.

There are striking differences as well as similarities between the three radioelements (Table III). The soil-to-roots transfer function shows a relatively fast rate constant for technetium and much smaller ones for plutonium and cesium. This pattern is consistent with measured field concentration ratios (tissue : soil) for these three elements. The radiocesium numbers ($10^{-3}$ to $10^{-4}$) reflect the difficulty in distinguishing and measuring changes in root content of trees growing in a heavily contaminated substrate such as the floodplain forest. In forests growing on sandy soils or soils lacking in clays, this function

could be much larger ($10^{-1}$). Upward translocation (roots to aboveground trees) is very rapid for radiocesium and varies with evapotranspiration rates during the growing season. Because of this variability rate, constants for this transfer were not calculated. Technetium also manifests rapid movement, whereas plutonium exhibits almost complete immobility.

Those transfers composing the return parts of the cycle reflect the differences between the nuclides as well as some site-related characteristics. Downward translocation is rapid for both technetium and cesium (0.25 vs 0.33) and is essentially nonexistent for plutonium. Leaf litter fall is the primary mechanism for the foliage-to-litter transfer. The high values for technetium and plutonium (0.97, 0.92) reflect the almost complete retention of these nuclides once they are translocated to the leaves, whereas in the case of cesium there is movement of radiocesium out of the leaves before they fall [15]. Leaching by rainfall (foliage to soil) reflects the mobility of cesium and has been demonstrated experimentally [15]. Rates of decomposition (litter to soil) can vary considerably between forests and are both species and climate dependent. The rapid transfers for cesium were derived from experiments [16] which tested leaf species of high nutrient content. The similar values for technetium and plutonium are a coincidence because they were arrived at by different calculational methods. Return of the radionuclides to soil from roots is an important part of the forest ecosystem biogeochemical cycle. Exudation occurs directly as part of the process of nutrient exchange between the plant and its soil substrate. The transfer coefficients shown in Table III reflect both root mortality, an important constituent of the turnover process, and estimates of exudation. Similarities in rates (0.23, 0.23, 0.15) are indicative of the overriding aspect of transfers through mortality as well as the considerable difficulty in obtaining direct measurements of nuclide exchange between root and soil. Physical resuspension is important only for plutonium, and in this humid forest ecosystem the process constitutes only a very small fraction ($10^{-5}$) of the cycle.

## Discussion

The environmental problems associated with contamination of land surfaces by long-lived radionuclides are once again a matter of public concern because of the Chernobyl accident. Questions about the treatment

and use of the affected land are being raised and evaluated in the countries involved. We have few published documents on the costs associated with the cleanup of contaminated land. Recently, and coincidentally, Baes, Garten, Taylor, and Witherspoon of Oak Ridge National Laboratory reviewed the long-term environmental problems of radioactively contaminated lands for the U.S. Federal Emergency Management Agency [17] and compiled a summary of costs associated with cleanup of contaminated land based on the limited data available. The costs range from about $3,000 to $24,000 (U.S. dollars) per hectare for various in situ treatment methods to hundreds of thousands of dollars per hectare for removal and disposal in an appropriate repository. Decisions on remedial actions, therefore, should take into consideration the benefits to be derived from a particular approach, with protection of the public from health hazards being the most important consideration.

How should severely contaminated land be dealt with? In this paper, I have illustrated the situation for temperate forests growing in areas where the soils have been contaminated with large quantities of fission products. Although the data are incomplete, they suggest, and the modeling results corroborate, the likelihood that the soil is, or will be, the sink for the radionuclides. Also, over time, decreasing quantities of the radioisotopes will enter the ecological cycles within the forest ecosystem. From our data, one can predict that temperate forests which have been contaminated by aerial deposition of radioisotopes will transfer these isotopes within a few years to the soil compartment where fixation processes will tend to make them unavailable. The time for such processes will depend on the mineralogical properties of the particular soils. Obviously, boreal and warm-climate forests differ drastically, and, as is well known, the boreal forest soils do not have the retention properties of temperate forest soils, and the local situations in those regions may require different approaches.

Nevertheless, forests offer a practical management alternative for contaminated lands under certain conditions. Forested lands that are heavily contaminated by aerial deposition may be best treated by being left alone and protected from fire and even from lumbering until safe levels have been reached through radioactive decay. Agricultural land which is too severely contaminated to use and which is not essential for crop production could be allowed to revert to forests either through natural succession or aerial seeding. This practice would provide

economic benefits from the land. Succession to a harvestable size ranges from 25 to 100 years. During this time, some radiological decay will occur. Additionally, closing of the forest canopy and establishment of an organic litter layer on the forest floor would tend to fix the radionuclides in place and enhance conditions for microbiological activity, which in turn would enhance biological fixation [17]. Additionally, forests would reduce radionuclide movement due to wind action and minimize remobilization through rain-induced erosion. On the other hand, fire can be a serious problem, which needs to be prepared for over a period of decades. Fire can cause resuspension of the contained radionuclides. We believe, therefore, that future land-management practices should take into account the positive attributes of forests as a landscape cover to minimize contamination problems, especially if this can be done within the framework of health protection needs and requirements for food production.

Acknowledgments

I am immeasurably grateful to my colleague Charles T. Garten, Jr., whose advice and assistance were invaluable in the preparation of this paper, and to B. Gordon Blaylock and F. Owen Hoffman for their encouragement and advice.

This work was sponsored by the Office of Health and Environmental Research, U.S. Department of Energy, under Contract No. DE-AC05-840R21400 with Martin Marietta Energy Systems, Inc. Publication No. 2817, Environmental Sciences Division, ORNL.

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Table I. Distribution of $^{137}Cs$ in a mixed deciduous forest approximately 30 years after soil contamination, based on data from Van Voris and Dahlman [3]

| Compartment | Mass ($g/m^2$) | Concentration (pCi/g) | Amount ( $Ci/m^2$) | Percent of total |
|---|---|---|---|---|
| Aboveground trees | | | | |
| Leaves | 478 | 27.9 | 0.013 | 0.0006 |
| Wood | 12 870 | 35.4 | 0.456 | 0.0208 |
| Stumps and roots | 5 348 | 350 | 1.89 | 0.086 |
| Litter | 500 | 20.6 | 0.010 | 0.0005 |
| Soil[a] | 260 000 | 8 439 | 2 194.1 | 99.89 |
| Total | | | 2 196.5 | 100.0 |

[a]Sample taken from 20-cm soil depth.

Table II. Estimated steady-state distribution of technetium, plutonium, and cesium among the various compartments of a deciduous forest

| Compartment | Percent of total system | | | |
|---|---|---|---|---|
| | Tc[a] | Pu[b] | Cs[c] | Cs[d] |
| Soil | 89.5 | 99.9 | 85.44 | 99.8 |
| Litter | 0.6 | 0.018 | 1.8 | 0.0005 |
| Tree stumps and roots | 1.4 | 0.071 | 4.2 | 0.08 |
| Aboveground trees | 8.0 | 0.0002 | 8.5 | 0.021 |
| Total | 99.5 | 100.0 | 99.94 | 99.90 |

[a]Values from Ref. 8.
[b]Values from Ref. 10.
[c]Values from Ref. 12. Turkey oaks on well drained, sandy soil with 5% clay (kaolinite); assumed root : aboveground distribution ratio was 1:3.
[d]Values from Refs. 3 and 4. Mixed deciduous forest growing on contaminated loamy clay soil with 24% clay (illite) (see Table I).

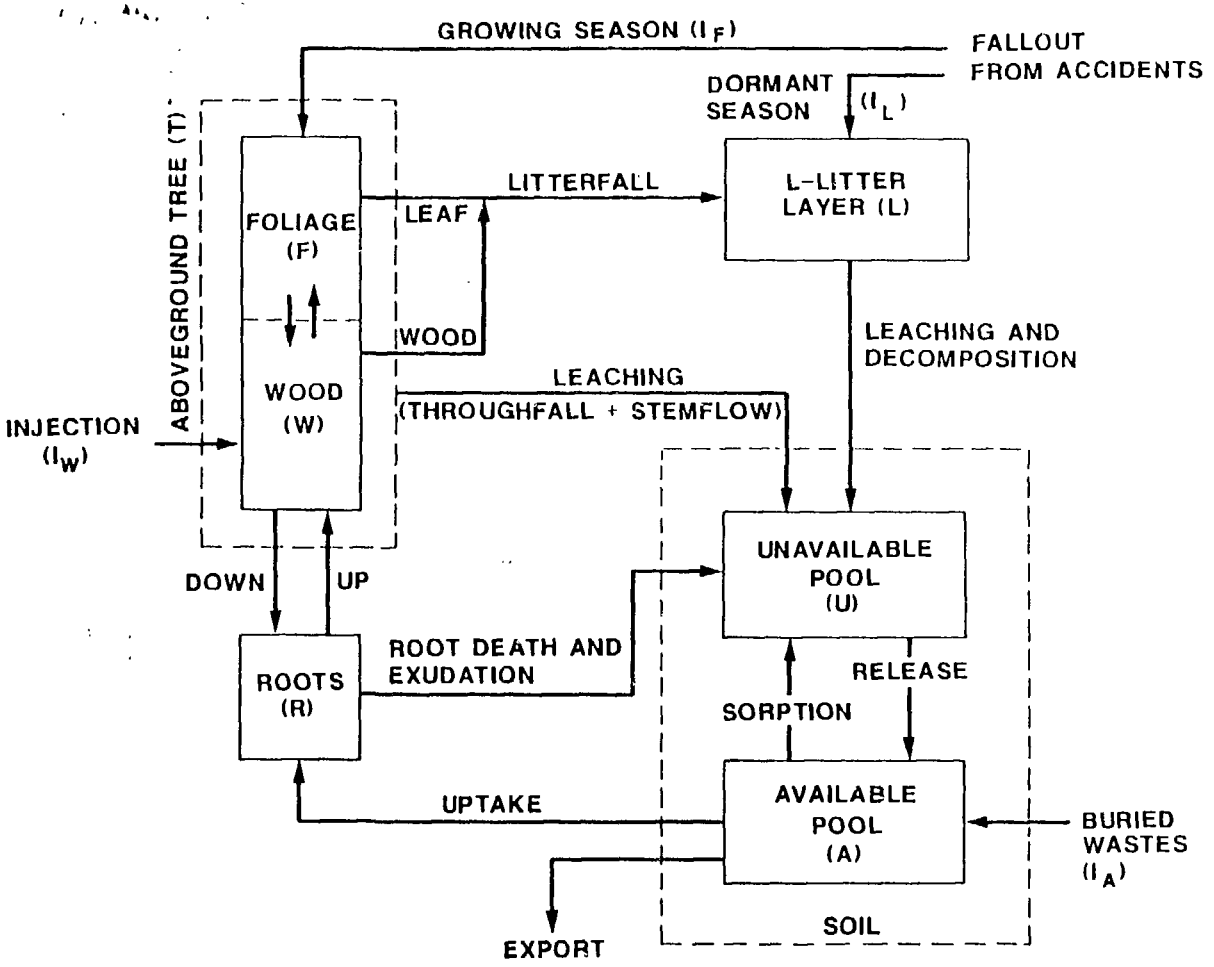
Table III. Estimated yearly rate constants for the cycling of plutonium, technetium, and cesium through a deciduous forest ecosystem

| Transfer | Description | Rate constant (per year)[a] Tc[b] | Pu[c] | Cs[d] |
|---|---|---|---|---|
| Soil to roots | Uptake by tree roots | 0.15 | 1.7E-04 | $10^{-3}$-$10^{-4}$ |
| Roots to above-ground trees | Upward translocation | 0.75 | 1.7E-04 | -- |
| Aboveground trees to roots | Downward translocation | 0.25 | Null | .33 |
| Foliage to litter | Leaf litter fall | 0.97 | 0.92 | 0.33-0.42 |
| Aboveground trees to litter | Wood litter fall | 0.0052 | 0.0092 | 0.002-0.004 |
| Foliage to soil | Leaching by rainfall | 0.004 | Null | 0.15 |
| Litter to soil | Decomposition and weathering | 0.47 | 0.48 | 1.3-3.5 |
| Roots to soil | Mortality and exudation | 0.23 | 0.23 | 0.15 |
| Soil to litter | Physical resuspension | NA | 8.5E-05 | NA |

[a]NA = not applicable.
[b]Values from Refs. 8 and 14.
[c]Values from Ref. 10.
[d]Values from Refs. 4, 15, and 16.

## FIGURE LEGEND

Fig. 1 Six-compartment, linear, donor-controlled model for simulating and analyzing radionuclide transfers within forest ecosystems. Entry of radioisotopes into the ecosystem can be through injection, fallout, accidents, or buried wastes. Source: C. T. Garten, Jr. Technetium-99 cycling in deciduous forests: Implications for radioactive waste management. Submitted to Environment International.


GROWING SEASON ($I_F$)
FALLOUT FROM ACCIDENTS
DORMANT SEASON
($I_L$)
ABOVEGROUND TREE (T)
FOLIAGE (F)
WOOD (W)
LITTERFALL
LEAF
WOOD
L-LITTER LAYER (L)
LEACHING AND DECOMPOSITION
INJECTION ($I_W$)
LEACHING (THROUGHFALL + STEMFLOW)
DOWN
UP
UNAVAILABLE POOL (U)
ROOTS (R)
ROOT DEATH AND EXUDATION
RELEASE
SORPTION
UPTAKE
AVAILABLE POOL (A)
BURIED WASTES ($I_A$)
SOIL
EXPORT