Radionuclide Fate and Effects

Naturally occurring and man-made radionuclides can be released into the environment at several stages in the nuclear fuel cycle. The releases may be planned or accidental, and both types can contaminate the environment. One goal of ecological research at PNL is to develop a basic understanding of how radionuclides behave in the environment, and to use this knowledge to build a scientific basis for making decisions about the siting, operation, and decommissioning of nuclear facilities and the methods for waste disposal.

The studies reported here deal with the full range of contaminant behavior and fate, from the initial physicochemical factors that govern radionuclide availability in terrestrial and aquatic environments to studies of contaminant transport by biological means. By design, we focus more on the biologically and chemically mediated transport processes and food-chain pathways than on the purely physical forms of contaminant transport, such as transport by wind and water.

A primary objective of the work is to develop realistic dose assessment models and to provide accurate parameters for applying those models. We use integrated laboratory and microcosm studies to define basic mechanisms; we use field studies to verify predictive models and to identify important processes at the community and ecosystem levels.

ENVIRONMENTAL TRANSPORT PROCESSES

Technetium is one of several radionuclides that could be released to the environment, primarily through the air. Recent research has shown that technetium is mobile in soils and is readily taken up by plants. These properties, combined with the element's long physical half-life, mean that humans could receive doses of technetium through food-chain transfer. Technetium's mobility in soil, which regulates its entry into the food chain, is influenced by pH, the presence of hydrous oxides of iron and manganese, the soil's organic matter content, and microbial activity. Technetium uptake by plants and observed toxicity in soil at levels >0.1 μg/g indicate that technetium behaves like a nutrient. Our prior laboratory studies with technetium and nutrient anions suggested that plants react to technetium much like they react to sulfur. To examine this hypothesis, we analyzed extracts from several plant species allowed to grow with their roots in contact with TcO_4^-. Our results showed that technetium is incorporated into higher molecular weight organic molecules, and that the degree of incorporation depends on the plant species and plant component.

The chromatographic behavior of the soluble technetium fraction after gel permeation, when extracted from the roots of soybean (Glycine max) and garlic (Allium sativum) plants, is shown in Figure 1. Most of the technetium present in soybean roots is soluble and still present in the TcO_4^- form. Garlic roots, however, contain only a minor fraction of technetium present as TcO_4^-; most of the technetium is incorporated into organic materials having higher molecular weight, which elutes faster from the gel column.

In all species of plants examined to date, the quantity of technetium transported to the portions of the plants exposed to the air (e.g., leaves and stems) was reduced whenever a significant fraction of technetium in non-TcO_4^- forms was incorporated in the roots. Also, animal feeding studies conducted this year show that technetium is principally concentrated in the yolk of Japanese quail eggs and in the hair of mammals. Yolks and hair are sulfur-bearing tissues, and the concentration of technetium in them supports our hypothesis that technetium is a sulfur analog.

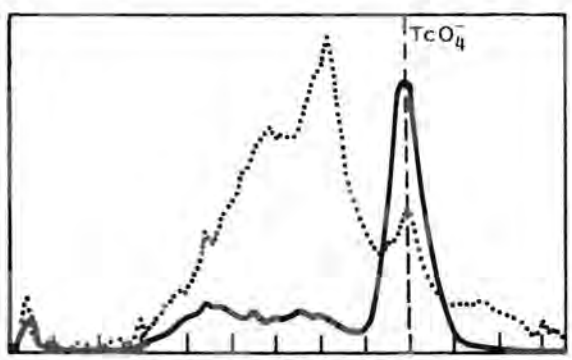

FIGURE 1. Technetium Is Incorporated into Several Organic Fractions Extractable from Plant Roots by Chromatography (see peaks at center). Anionic technetium (peaks at right) is ten-fold higher in soybean roots (solid line) than it is in garlic roots (dotted line).

In addition to our technetium work, we are studying the role of transuranic elements in food-chain transfers. Previous studies with plutonium and neptunium show that the roots of soybean and alfalfa plants absorb these elements and store them in their metabolic compartments. Plutonium supplied as $Pu(NO_3)_4$ to hydroponically grown plants is poorly absorbed by roots; less than 0.001% is transferred to shoot tissues. Root absorption can be increased by three orders of magnitude by using Pu_3DTPA_3 to maintain solution solubility. This treatment also increases the plutonium fraction transferred to shoot tissues. Most of the plutonium metabolized by roots and leaves is found in the MW fraction of solubles >10,000. In our experimental soybean plants, only 0.00035% of the absorbed plutonium was in the seed. Most of this (>80%) was unsolubilized in the seed coat and would be unavailable for absorption if ingested by animals.

Our experiments with neptunium show that it behaves differently than plutonium. In soybean, alfalfa, and more recently, bushbean plants, absorption is nearly total; shoot tissues contained ~30%, 6%, and 40%, respectively, of the root-absorbed neptunium. Unlike plutonium, metabolized neptunium was distributed among several high molecular weight fractions, e.g., >30,000, 30,000-5000, 5000-500 and <500. This was found in leaves, stems, and roots. For all plants studied, less than 5% of the neptunium was found in stem tissues, suggesting the presence of efficient plant solubilization processes. Neptunium was also found in seeds at maturity in bushbean plants. A substantially higher fraction was observed than that found with plutonium. Most of the seed-incorporated neptunium is soluble and extractable (~75%).

The soil processes that keep transuranic elements soluble prior to root absorbtion appear to influence greatly plant uptake and, therefore, the movement of these elements into the ingestion pathway (Figure 2). Differences in uptake among plants and the fractional distribution of plutonium and neptunium among component plant parts (seed, leaf/stem, root) reflect the fact that some crops (cereal grains, roots or tubers, leafy vegetables or forage) contribute more of certain elements to the food chain than others.

In addition to our laboratory studies, we are also conducting field studies with plutonium, americium, cesium, neptunium, and uranium. The need here is to determine what differences from the laboratory data can be expected as plants are exposed for long periods of time under realistic environmental conditions. The experimental method involves agricultural and range plant species in contaminated outdoor soil columns known as lysimeters. We expect the responses to vary with temperature and moisture fluctuations, cropping schedules (for legumes, cereal grains, and grasses), and the type of tillage operation used. Annual progress involves maintaining the lysimeter system, recording climatic variables, and tilling, seeding, irrigating, weeding, and harvesting the crops.

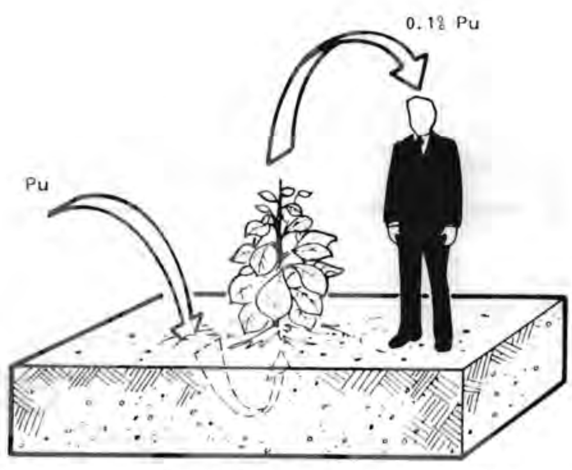

FIGURE 2. Most of the Plutonium Entering the Soil Remains Bound to the Soil Fraction, and the Amount Entering Man Through Food Chains is Small. The biologically available fraction entering man, however, can vary over several orders of magnitude depending on microbial and other soil processes.

Currently, comparison of ^{239}Pu uptake for peas and barley shows that the uptake of nitrate ^{239}Pu was 7 to 20 times greater than uptake of the oxide form. However, the uptake of both chemical forms decreased by about an order of magnitude over three growing seasons. This event would not be readily ascertainable by laboratory approaches. We also expect to differentiate the more important environmental and culture variables that influence contaminant availability.

RADIONUCLIDE TRANSPORT PARAMETERS

Radionuclide transport pathways are not known with certainty for many elements. Our work on transport pathways has permitted us to identify areas where clarification is most urgently needed. We recently conducted laboratory experiments on Japanese quail to estimate the transfer from feed to eggs and meaty tissues (Figure 3). The magnitude of this transfer is considerably greater (up to three

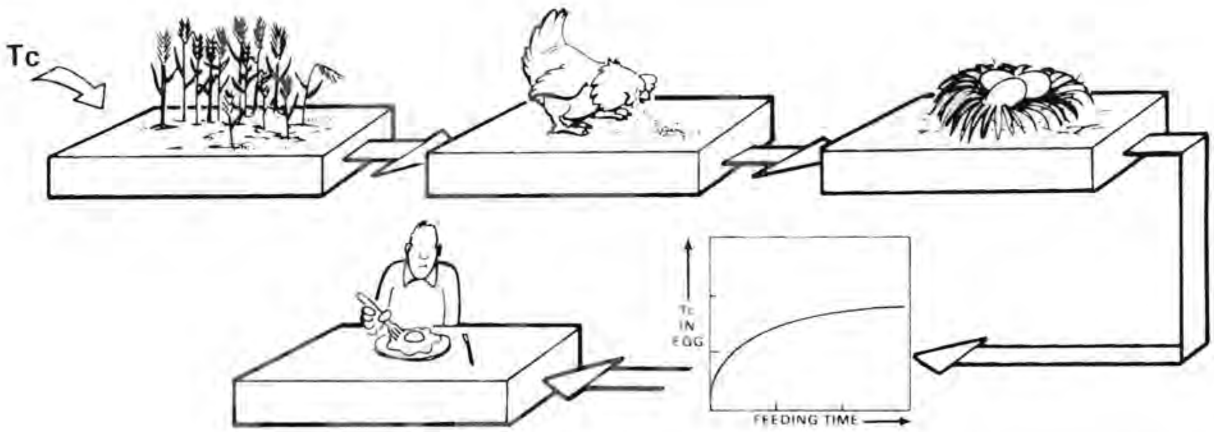

FIGURE 3. Eggs Proved to be an Unexpected Point of Accumulation for Technetium, and As Such Could Provide a Source of Contamination for Man. Technetium is probably a sulfur analog (see *text*).

orders of magnitude) than previously believed. This accumulation may be explained by the fact that technetium behaves like sulfur in biochemical pathways. We expect that these data will result in upward revisions of commonly used technetium transfer coefficients for the feed-to-fowl ingestion pathway. Revisions to calculated ingestion doses for man will follow.

Our work this year also involved reviewing the parameters used in dose assessment models for calculating the dose that humans would receive if they consumed fish from contaminated waters. Both abiotic and biotic factors have been under examination. We are seeking a general relationship for use in predicting the radionuclide concentrations in fish in different contaminated environments. Factors reviewed for the relationship include physicochemical speciation, the influence of stable isotope dilution, the presence of chemical analogs, and the known biological role of the radionuclide. Approximately 30 radionuclides common to the nuclear fuel cycle are included in this study. We expect this work to result in a reference volume or handbook for use in dose assessment.

ECOLOGICAL INTERACTIONS INFLUENCING REMEDIAL ACTIONS AT WASTE SITES

Radionuclides disposed of in shallow-land burial sites may be moved by root intrusions and animal burrowing. The extent to which biotic transport occurs depends on the type of plants and animal communities at a site. To determine the differences, we examined the ocurrence of pocket mice, a common burrowing animal, in areas primarily vegetated by either cheatgrass or wheatgrass. These two grasses can be used to stabilize the soil surface of low-level waste burial grounds in western climates. Our results show that pocket mice were five times more abundant in wheatgrass-dominated communities than in cheatgrass areas. We then calculated the relative radionuclide concentrations at the soil surface in each community and found that concentrations were higher and increased for a period of 70 to 100 years in wheatgrass communities because of the burrowing activity of these animals (Figure 4).

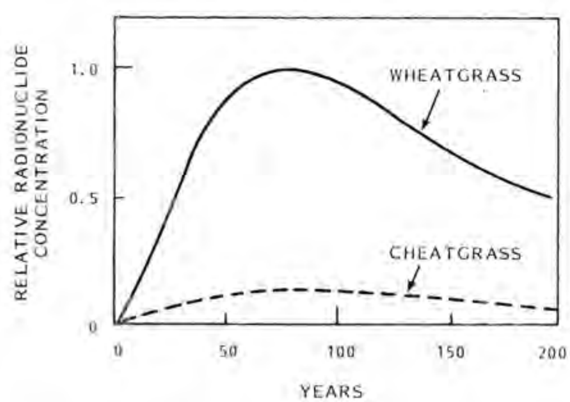

FIGURE 4. Burrowing of Small Animals Makes Radioactivity More Available to Grasses Growing on the Soil Surface. When burrowing activity is projected through a 200-year interval (using models developed in this program), we found that certain grasses favor transport of radioactivity to the surface.

The contribution of these biotic transport processes to radionuclide movement has yet to be evaluated quantitatively. The test results do imply that the choice of one surface cover over another can influence both the long-term biotic transport potential of radionuclides and their resultant dose to populations.

Field work on how to clean up inactive uranium mills entered its third season this year. Our goals have been to identify and develop methods for stabilizing the surface of tailing piles, and to find a way of preventing plants and animals from penetrating surface covers, thus allowing radon gas to escape. Field data obtained from the vicinity of inactive uranium mill tailings sites were grouped using the clustering technique into six general plant community types: steppe, annual steppe, shrub steppe, woodland, forb-dominated steppe, and salt-shrubs. The grouping will now help us identify the most important environmental factors that affect protective covers.

We conducted model simulations and field lysimeter studies of rock and plant covers to evaluate the effect that surface cover type has on moisture dynamics within the overburden and underlying tailings. The results of the simulations and the preliminary findings of the lysimeter studies show that rock covers increase the moisture content of soil by reducing surface runoff and evaporation. If a rock cover is applied to tailings in such a way that plant growth is eliminated, water will accumulate in the tailings and eventually percolate into the ground water. Soluble contaminants in tailings could thus contaminate groundwater.

A major product of this year's work is a plan for designing covers that satisfies the needs of a particular site (Figure 5). A designed cover must control surface erosion adequately and not jeopardize the containment of the biologically toxic components of the tailings. Adequate erosion control is, logically, that which is required to keep the protective layer intact for the design life of the containment system. A surface cover, therefore, can be any combination of soil, rock, and vegetation that is effective and economical.

A slow-release herbicide in a polymeric carrier was developed and tested during the past year. Our goal was to create an effective system for controlling plant root penetrations through the earthen covers used to cap uranium mill tailings piles. A polymer system containing 58% polymer, 24% herbicide, and 18% carbon black was developed and tested. Our studies show that the minimum effective concentration of the herbicide in soil for our delivery system was in the range of 0.3 to 6.4 ppm for the species tested.

Loose rock, asphalt, and multilayered earthen barriers were tested to see if they could withstand the burrowing of townsend ground squirrels (Spermophilus townsendii) and white-tailed prairie dogs (Cynomys leucurus)--information that simply has not been available. Each barrier (Figure 6) was placed in two of eight outdoor pens; the remaining pens served as controls. The ground squirrels and prairie dogs burrowed a maximum of 15 cm into the rock layer of the mulitilayer earthen barriers. The deepest penetrations in all rock layers occurred along the walls of the boxes. No evidence of penetration by prairie dogs was found with the asphalt barriers. We found that nearly half of the soil was displaced by burrowing, but there were few distinct tunnels. The 3-inch-thick asphalt barriers were effective in preventing intrusion by both rodent species tested. The larger of the two animals, the prairie dogs, were able to move the rock more easily, but were still excluded by the barriers. This finding implies that intrusions into buried waste (uranium mill tailings in this case) by plants and animals can be effectively deterred by an adequately designed barrier system.

TRANSURANIC CHEMICAL SPECIES IN GROUNDWATER

The migration of mobile forms of the transuranic radionuclides in groundwater is being studied by a resin bed sorption technique. Previous studies at this site showed that traces of plutonium are transported in the groundwater in a soluble, anionic chemical form in which the plutonium is predominantly (~90%) in the V or VI oxidation state. A hydroxy carbonate anionic complex of the plutonium may be this mobile form. During fiscal year 1982, we installed a series of three new sampling wells. Once the mobile radionuclide chemical species form at the crib/trench, they pass through the soil column relatively unattenuated in the groundwater and do not accumulate in downstream soils.

ANALOGS FOR TRANSURANIC CHEMISTRIES

The goal of this research has been to develop quantitative understanding of the environmental behavior of naturally occurring rare earth elements so that the data might be used to estimate the behavior of some of the transuranic elements. Attention has been focused on americium and curium--transuranic

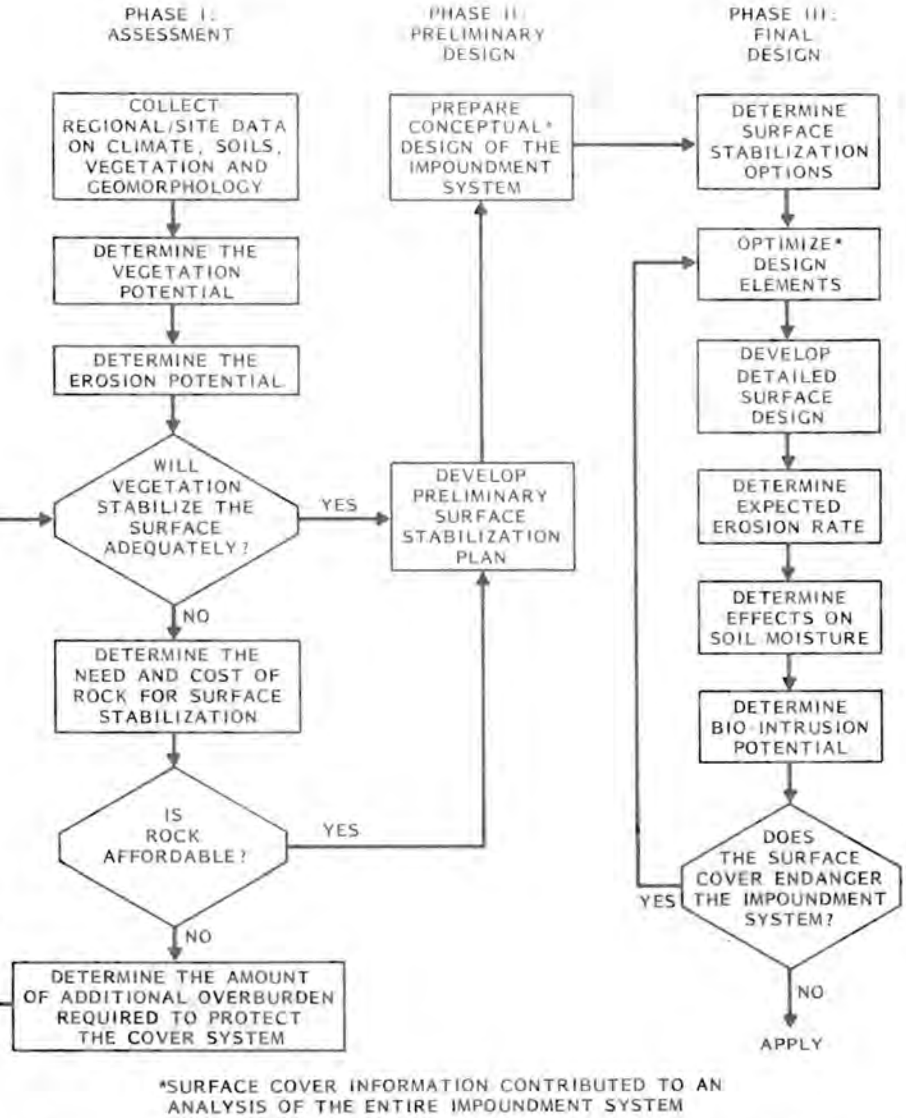

FIGURE 5. When Applying Ecological Data to a Surface Stabilization Problem, Many Detailed Considerations Must Be Systematically Evaluated. A significant product of the current year's work was development of the model shown above, which is designed to protect a containment system from erosion and biotic intrusion.

elements that also exist environmentally in the +3 oxidation state.

Data on rare earth element behaviors are being investigated for their effectiveness in estimating the radiological dose that man would receive from environmentally weathered transuranic elements. An important consideration in estimating human dose regards the concentrations of the natural rare earth elements in food products. If the hypotheses are correct, these rare earth concentration data might be useful in estimating the potential dietary intake of transuranic elements which have weathered in the environment for long time periods. Foodstuffs, representing a typical "market basket" for the American populace, were obtained, and sub-samples of the edible inner portions were analyzed by radiochemical neutron activation analysis for the entire suite of rare earth elements. In general the compiled data demonstrate that the rare earths are present in foodstuffs at extremely low levels, near the detection limits of our state-of-the-art analytical methods (parts per billion). Concentrations in foods represent depletions of approximately 10^{-5} from the total soil concentrations based on average crustal abundance.

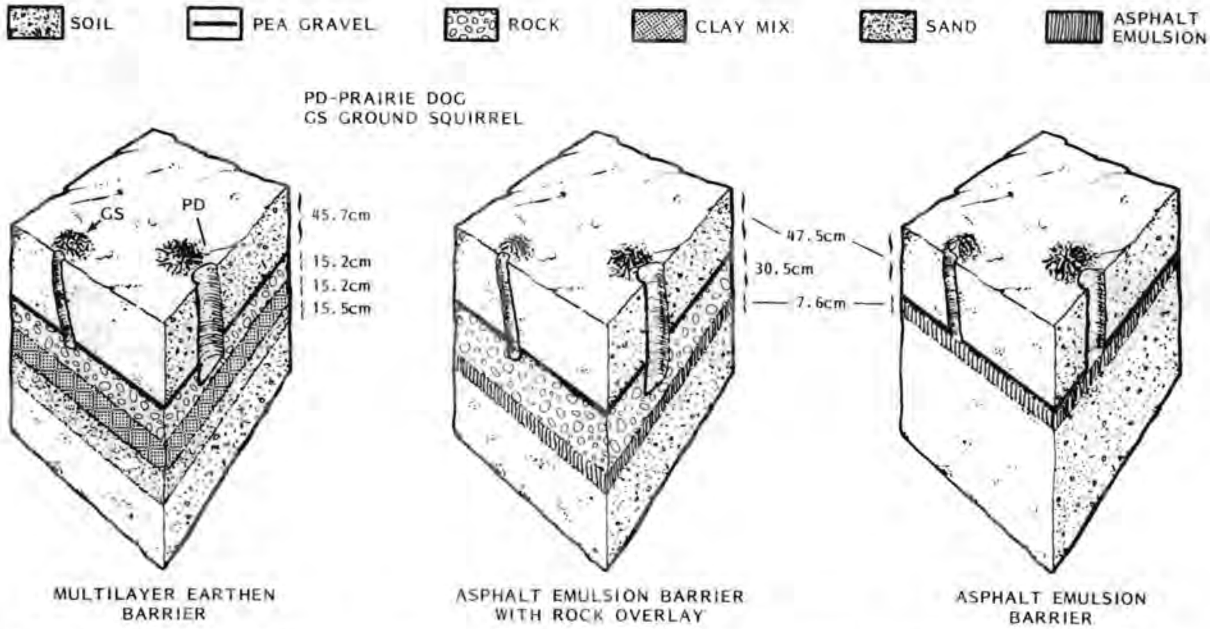

FIGURE 6. Information Has Not Previously Been Available on Practical Techniques to Prevent Animal Intrusion at Waste Burial Sites. Studies to optimize several different types of a barrier system showed that a properly configured asphalt layer was highly effective. Animals could penetrate rock layers, but did not burrow through the asphalt layer.

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