Quoted

THE ROLE OF PHYSICAL PROCESSES IN THE TRANSPORT OF MAN-MADE RADIONUCLIDES IN ARID ECOSYSTEMS

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BACKGROUND

Studies on the ecological behavior of man-made radionuclides in arid ecosystems began in the early 1940's with the development and testing of the worlds first atomic bomb at the Trinity Site in south-central New Mexico. Rather intensive radioecological studies on plutonium and other radionuclides were conducted soon after the test in the desert surrounding the Trinity Site by the University of California at Los Angeles (Larson et al., 1951, Leitch, 1951). In the 30 yr following this historic nuclear weapons test, a tremendous quantity of data was gathered on the distribution and transport of fission and activation products deposited as fallout as a consequence of measurable accumulation and concentration of some fallout radionuclides in natural and human food webs (Hanson, 1967; Pendleton et al., 1964). It was not until the mid 1970's and especially the early 1980's that intensive radioecological studies on plutonium and other actinides began in the U. S.

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under the auspices of the DOE, Office of Health and Environmental Research (US DOE, 1980) and Nevada Applied Ecology Group (US DOE, 1985) research programs.

Most of the radionuclide data summarized in this section were collected at the Nevada Test Site, Trinity Site (New Mexico), Los Alamos National Laboratory (New Mexico), Rocky Flats Plant (Colorado), Idaho National Engineering Laboratory and Hanford Reservation (Washington state). All of these sites average less than 500 mm of annual precipitation. Several, including Nevada Test Site, Trinity Site, Hanford Reservation and Idaho National Engineering Laboratory average less than 220 mm per year.

RADIONUCLIDE DISTRIBUTION IN ARID ECOSYSTEMS

Spatial and Temporal Relationships

Most sources of radionuclides presently in arid ecosystems in the U. S. were initially deposited on the ground surface decades ago as single contamination events or as chronic events over long periods of time. Results from field studies in arid and semi-arid areas in the western U. S. show quite convincingly that soil will eventually be the primary repository of plutonium, cesium, strontium and most other radionuclides with long physical half-lives (i.e. yrs-millennia) (Watters et. al, 1983; Little, 1980, Hakonson and Nyhan, 1980, Ritchie and McHenry, 1990; Gilbert et. al, 1987; Romney et al., 1987). Furthermore, the importance of soil as a reservoir for many radionuclides is often independent of source term (e.g. weapons fallout, effluents, accidents) and the type of terrestrial ecosystem (e.g. deserts, woodlands, shrublands, grasslands) (Watters et al., 1983). For example, on a unit area basis, the percentage of the total plutonium inventory associated with the soil component

of contaminated ecosystems was over 99% at four different arid sites contaminated with different sources of plutonium (Watters et al., 1983). Less than one percent of the inventory was associated with biological components of these ecosystems (Watters et al., 1983).

After the contaminating events have ceased, the concentration of radionuclides on the soil surface have usually decreased, as shown by the plutonium data in Trinity Site soils (Table 1), due to horizontal and vertical redistribution and, in the case of shorter lived ^{137}Cs and ^{90}Sr, also to radioactive decay. Note that the concentrations of plutonium in the 0 - 2.5 cm soil profile at Trinity Site, (where average annual precipitation is about 20 cm) decreased by a factor of 10 over a 23 year period. Corresponding data on ^{90}Sr and ^{137}Cs in surface soils from Nevada Test Site (about 13 cm annual average precipitation) have shown less dramatic reductions (i.e. up to a factor of two) (Romney et al., 1983). However, the soil surface in many areas of the test site is protected with erosion pavement, a natural stone covering, that greatly reduces the potential for horizontal movement of radionuclides by erosion processes.

As radionuclides move into the soil, they can become less available for transport by wind and water erosion. For example, in the stream channels used for treated radioactive liquid waste disposal at Los Alamos, only 4-20% of the plutonium in channel alluvium was in the 0 - 2.5 cm depth profile where it could be readily transported to downstream areas by storm runoff (Table 2). Barring major down-cutting through the alluvium during extreme runoff events, the plutonium that was deeper in the profile would likely continue to remain unavailable for transport by runoff processes.

Table 1. Comparison of Plutonium Concentrations in Surface (0 to 2.5 cm) Soils from Chupadera Mesa as a Function of Time After the Atomic Bomb Test at Trinity Site in 1945 (from Hakonson and Nyhan, 1980).

Plutonium concentration, nCi/m^2		
1950	1951	1973
746(0.31)* n = 6	341(0.82)* n = 3	18(0.48)* n = 8

*Parenthetic value is coefficient of variation (standard deviation/mean).

Table 2. Percentage of Plutonium Inventory As A Function of Depth In Alluvial Soils At Los Alamos, New Mexico (Adapted from Hakonson and Nyhan, 1980).

Depth, cm	Mortandad Canyon[1]	Acid-Pueblo Canyon[2]
0 - 2.5	20(0.44)[3]	4.0(0.76)
2.5 - 7.5	36(0.23)	10(0.48)
7.5 - 12.5	22(0.55)	20(1.3)
12.5 - 30	22(0.79)	66(0.18)

[1]was actually receiving treated radioactive liquid waste.

[2]had not received waste for 10 years.

[3]mean percent (n = 10) and coefficient of variation ($\sigma/\bar{x}$).

In another example from the fallout areas of the Nevada Test Site, 30-60% of the ^{90}Sr and ^{137}Cs inventory in soil, 18 years after atmospheric testing ceased, was associated with depths below 2.5 cm (Romney et al., 1983). Barring major disturbances of these soil profiles at the test site, both ^{90}Sr and ^{137}Cs will likely remain in place below the ground surface until radioactive decay has occurred.

Particle Size Relationships

An especially important distributional relationship that influences radionuclide transport in arid sites is the one between contaminant concentration and soil particle size. The highest concentrations of many radionuclides in soil are usually associated with the smaller particle size fractions (Watters et al., 1983; Little, 1980; Tamura, 1975; Nyhan et al., 1976; Essington and Romney, 1986) as illustrated in Table 3. For example, in liquid effluent receiving areas at Los Alamos, highest concentrations of plutonium and cesium-137 in intermittent stream bed alluvium are invariably in the silt-clay (< 53 μm) fraction.

There are exceptions to this relationship as shown for plutonium in surface soil from ground zero (Area GZ) at Trinity Site (Table 3). Highest plutonium concentrations (5.3 pCi/g) were found in the sand size fraction (1-2 mm) while very low concentrations (< 0.1 pCi/g) were measured in the < 53 μm fraction. Similar results were also obtained from nuclear ground zero sites at Nevada Test Site (Lee and Tamura; 1981) and have been attributed to the deposition of larger plutonium particles closer to ground zero than further along the fallout pathway.

Table 3. Comparative Distribution of Plutonium in Silt-Clay Fraction (< 53 μm) of Surface Soil From New Mexico (adapted from Hakonson and Nyhan, 1980).

	Los Alamos		Trinity Site	
	Mortandad Canyon	Acid-Pueblo Canyon	Area GZ	Area 21
pCi/g[1]	1500	85	0.07	3.8
Soil Weight (%)	2.2	3.0	8.9	36.0
Pu in Fraction (%)	14.0	7.0	0.78	73.0

[1]Plutonium concentration in 0 - 2.5 cm depth profile.

Even though concentrations of radionuclides may be much higher in a particular soil size fraction, the total inventory of radioactivity in that fraction will depend on its mass relative to the whole soil. For example, despite the higher concentrations of plutonium in the silt-clay fraction of Los Alamos alluvial soils, less than 15% of the total plutonium inventory in whole soil was in this fraction (Table 3). In contrast, the silt-clay fraction in Area 21 at Trinity Site (a rangeland site) not only exhibited the highest plutonium concentrations, but it also contained over 70% of the total plutonium inventory in soil (Table 3).

Soil will serve as the primary reservoir of many long-lived radionuclides in arid environments and complex spatial, chemical, and physical relationships will determine the mechanisms of transport and their relative importance. Wind and water sort soil particles during the detachment, transport, and deposition phases of erosion. Because radionuclide concentrations can be strongly related to soil particle size, there is a potential to enrich concentration of the radionuclide in eroding soil. This potential for enrichment, can affect the long term redistribution of radionuclides and their transport to biological components of ecosystems.

ABIOTIC PROCESSES

The importance of wind and water in transporting many radionuclides in arid environments results from the fact that most radionuclides deposit near quantitatively in soil and are tightly bound to this matrix (Hanson, 1975; Ritchie and McHenry, 1990; Whicker and Schultz, 1982). Consequently, processes which transport soil also transport soil-associated contaminants.

In many cases, both wind and water preferentially detach and transport the finer size fractions that often contain the highest concentration of the radionuclide. Moreover, the finer soil fractions are carried farther (and deposited later) than coarser fractions of eroding soil. Recent advances in the ability to predict plutonium transport by hydrologic processes is discussed here to illustrate relationships that govern wind and water transport of radionuclides. More detail on wind erosion and radionuclide transport by wind can be found in Bagnold, 1941; Graf, 1971; Marshall, 1973; Gallegos, 1978; Sehmel, 1980; and Anspaugh et al., 1974.

Hydrologic processes of particular importance in the physical transport of soil-associated radionuclides include soil detachment by raindrop splash (Ellison, 1945; Mutchler and Young, 1975; Martinez-Menez, 1979) and soil detachment and transport by overland flow (Meyer and Wischmeier, 1966; Foster and Meyer, 1972; Wischmeier and Smith, 1978). Soil particles detached by raindrop impact are important because they can be deposited on vegetation surfaces and thus provide a pathway for movement of soil-associated radionuclides to animals (see section on biotic processes). Sediment transported by overland flow is important because it can redistribute contaminants within a watershed and also deliver them to stream channels for subsequent transport to downstream areas.

The combined phases of runoff, soil erosion, sediment transport, and deposition of sediment on upland areas and in stream channels usually result in enrichment of smaller soil particles and organic matter in transported sediment (Graf, 1971) including concentration of sediment associated contaminants (Massey and Jackson, 1952; Menzel, 1980; Lane and Hakonson,

1982). This enrichment is often expressed as an enrichment ratio, defined as the concentration of contaminant in the transported sediment divided by its concentration in the residual or uneroded soil. Enrichment ratios have been related to sediment concentration, sediment discharge rate, and sediment yield (Massey and Jackson, 1952; Menzel, 1980). Lane and Hakonson (1982) analyzed sediment transport rates by particle size classes in alluvial channels and derived the following expression:

$$ER = \frac{\Sigma[Cs(di) \cdot Qs(di)]}{Cs\ \Sigma[Qs(di)]} \qquad (1)$$

where:

ER = alluvial channel enrichment ratio,

Cs(di) = Concentration of contaminant in sediment particles of size class i, with representative diameter, di, in millimeters.

Qs(di) = Sediment transport (mass/time) for particles in size class i, with representative diameter, di, in millimeters.

Cs = Mean concentration of contaminant in soil over all particle size classes.

Equation 1 supports the empirical observation that enrichment ratio increases with decreasing sediment discharge rates. For example, at very low sediment discharge rates (those associated with low runoff velocities) the bedload discharge rate for coarse sediment particles is low and most of the transported sediment is in the smaller particle size classes. Under such conditions, ER would approach the ratio of concentrations in the finest size classes (Cs(di)) to the mean concentration over all size classes (Cs). At

high sediment discharge rates (those associated with high runoff velocities) more of the bed sediments are in transport. At the extreme, if all of the bed sediments were in transport in the same proportion as they exist in the bed material, ER in Equation 1 would be unity.

Field measurements of enrichment ratios for nutrients and plutonium at several locations in the United States are listed in Table 4. The first four entries are for soil nutrients in runoff from small agricultural areas; mean values vary from 2.6 to 7.1. The next three entries represent enrichment of fallout plutonium in runoff from agricultural watersheds; mean valucs range from about 1.6 - 2.5. The last entry represents enrichment of plutonium in runoff in ephemeral stream channels at Los Alamos, New Mexico. Field derived values for Los Alamos ranged from 1.4 to 13.3 with a mean of 5.5. Predicted enrichment ratios for Los Alamos stream channels [Eq. (1)] ranged from 2.9 to 7.0 with a mean of 5.2 (Lane and Hakonson, 1982). The close agreement between observed and predicted enrichment ratios suggests that particle sorting alone can account for ratios observed at Los Alamos. Although other factors undoubtedly influence the observed enrichment ratios, several analyses (Menzel, 1980; Foster et al., 1981; Lane and Hakonson, 1982) suggest that particle sorting alone can account for observed enrichment ratios. In spite of wide differences in watershed size, hydrologic regime and chemical characteristics, the enrichment ratios resulting from sediment transport given in Table 4 are quite similar for several sediment associated chemicals. Particle sorting is clearly one of the important factors involved in transport of soil and sediment associated radionuclides.

ıble 4. Approximate Enrichment Ratios for Nutrients and Plutonium Associated with Sediment at Various ›cations in the United States.

Land use nd location	Approximate enrichment ratios mean	range	Comments	References
opland, USA[a]	4.5	2.5 - 7.4	Nitrogen	Menzel, 1980
	3.6	2.6 - 6.0	Phosphorus	Menzel, 1980
ngeland, USA[a]	2.6	1.1 - 6.7	Nitrogen	Menzel, 1980
	7.1	2.7 - 17	Phosphorus	Menzel, 1980
opland, USA[b]	1.6	1.1 - 2.5	Fallout Plutonium	Muller et al., 1978
sture, USA[b]	2.3	0.8 - 4.0	Fallout Plutonium	Muller et al., 1978
ked Cropland, USA[c]	2.5	1.2 - 4.0	Fallout Plutonium, Transport in Perennial River	Sprugel & Bartelt, 1978
niarid, USA[d]	5.5	1.4 - 13.3	Waste Effluent Plutonium Transport in Ephemeral Streams	Lane & Hakonson, 1982

ıall agricultural watersheds (5.2 - 18 ha) at Chickasha, Oklahoma.
ıall agricultural watersheds (2.6 - 2.9 hg) near Lebanon, Ohio.
·eat Miami River (Drainage area = 1401 km^2) at Sidney, Ohio.
›s Alamos Watersheds (176 - 15,000 ha) near Los Alamos, New Mexico.

BIOTIC PROCESSES

Transport to Vegetation

Two mechanisms for transport of soil contaminants to terrestrial plants are absorption by roots and deposition of contaminated soil particles on foliage surfaces with or without subsequent absorption into plant tissues. Despite the host of chemical, biological and physical factors which can modify the physiological (chemical) availability of radionuclides and subscquent transport to internal plant tissues (Wildung and Garland, 1980; Adriano et al., 1980; Cataldo and Vaughn, 1980), field studics suggest that contamination of foliage surfaces with soil particles containing the radionuclides is a major transport mechanism under many arid site and radionuclide source conditions (Romney and Wallace, 1976; Romney et al., 1987; White et al., 1981; Arthur and Alldredge, 1982). For example, comparative studies of plant uptake of plutonium under both field and laboratory conditions generally yield results of the type shown in Table 5. Laboratory studies focused on root uptake of plutonium from soils yield concentration ratios which are at least one order of magnitude (and often 2-3 orders of magnitude) lower than ratios observed under comparable conditions at field sites. The differences in concentration ratios between laboratory and field studies implies that a mechanisms exists in arid environments for delivering at least 10 times more plutonium to vegetation than would be predicted based upon greenhouse studies. The higher ratios observed at field sites are generally attributed to the presence of surficial contamination on field site vegetation (Romney et al., 1987; Hakonson and Nyhan, 1980; Little et al., 1980). That conclusion is supported by the obvious presence of soil on foliage surfaces in the field and

Table 5. Comparison of plutonium concentration ratios for field and glasshouse conditions (Romney and Wallace, 1976).

Soil Source	Field	Glasshouse
NTS[a] Area 11B	1.3×10^{-2} to 1.6×10^{-1}	1.5×10^{-4}
NTS Area 11C	4.5×10^{-2} to 3.4×10^{-1}	1.8×10^{-4}
NTS Area 13	7.8×10^{-2} to 4.4×10^{-1}	1.1×10^{-4}

[a]NTS (Nevada Test Site).

by the ability to remove up to 90% of the plutonium contamination from vegetation by washing (White et al., 1981; Arthur and Alldredge, 1982).

Studies at Los Alamos demonstrated that rain-splash of soil particles with subsequent deposition on foliage surfaces can easily contribute all of the plutonium measured in field-site vegetation (Dreicer et al., 1984). More importantly, those studies, which employed a labeled-soil particle technique and the scanning electron microscope, have shown that relationships that govern translational movement of plutonium by soil erosion processes also govern transport of plutonium to foliage surfaces. For example, the energy of impacting raindrops caused an enrichment of the smaller soil particles (up to 105 μm in this study) on foliage surfaces. The amount of soil deposited on the plants was also related to height from the ground surface and characteristics of the rainstorms. Calculations based on the mass and plutonium content of soil measured on the plants demonstrated that the rainsplash mechanism could easily account for the observed plant/soil concentration ratios of 5×10^{-2} (White et al., 1981; Foster et al., 1985). While absorption of plutonium through leaf surfaces has been demonstrated (Cataldo and Vaughn, 1980) it is likely to be of limited importance in arid field sites particularly for annual or deciduous vegetation.

Studies on the uptake of plutonium by vegetable crops grown in field sites at Los Alamos show that as much as 50% of the plutonium in crop samples was surficial contamination that could be removed by washing (White et al., 1981). Plutonium that cannot be removed from vegetable crop surfaces in arid environments does not necessarily reflect contamination of plant tissues. Cataldo and Vaughn (1980) showed that submicron particles on foliage surfaces are difficult to remove by either simulated wind or rain.

TRANSPORT TO ANIMALS

As with vegetation, soil can be a major source of radionuclides to animals in arid ecosystems. Soil particles can be transported to animals in association with exterior surfaces of food and by direct transfer of soil to the animal via inhalation, ingestion and contamination of the pelt.

Plutonium is the best example of a radionuclide whose transport to animals in arid ecosystems is dominated by physical processes. Data from many field sites and source conditions show that gut availability of plutonium in a variety of animals including rodents, deer and cattle is very low leading to very low concentrations of plutonium in internal tissues and organs (Smith, 1977; Moore et al., 1977; Hakonson and Nyhan, 1980; Arthur et al., 1987; Romney et al., 1970). Highest concentrations are generally found in tissues exposed to the external environment including the pelt, gastro-intestinal tract and lungs. At Los Alamos, about 96% of the plutonium body burden in rodents from the canyon liquid waste disposal areas was in the pelt and gastro-intestinal tract (Hakonson and Nyhan, 1980).

Because soil passes through the gastro-intestinal tract of free-ranging animals on a daily basis, there is a potential to redistribute soil radionuclides across the landscape. Studies at Nevada Test Site with cattle (Moore et al., 1977), at Rocky Flats Plant with mule deer and small mammals (Little, 1980; Arthur, 1979), and at Idaho National Engineering Laboratory with small mammals and coyotes (Arthur and Markham, 1983; Arthur et al., 1980) demonstrate that horizontal (and vertical, in the case of burrowing animals) redistribution of soil plutonium does occur as animals move within and outside contaminated areas. While the magnitude of this transport is considered small

over the short-term, there are circumstances where it assumes much more importance. For example, in a nuclear waste burial site at Hanford, defecation of ^{90}Sr on the ground surface and surrounding area by jackrabbits burrowing into the waste, required remedial action to prevent further migration of the strontium (O'Farrell and Gilbert, 1975).

Strontium and cesium transport to animals in arid ecosystems involves a combination of physical and physiological processes. The more tightly bound these radionuclides are to soil (related to clay content of soil), the more their transport will be governed by soil particle transport. Data on ^{90}Sr and ^{137}Cs in small mammals from Nevada Test Site (Romney et al., 1983) and Idaho National Engineering Laboratory (Arthur et al., 1987) often show relatively high concentrations in lung, pelt and gastro-intestinal tract with similar or much lower concentrations in internal tissues and organs.

CONCLUSIONS

In arid U. S. ecosystems, contaminated with radionuclides several decades ago, nearly 100% of the residual radioactivity is sorbed to the soils and sediments of receiving areas. For radionuclides that are tightly bound to soils, such as plutonium, erosional processes, driven by wind and water, control horizontal movement of these radionuclides as well as their entry into biological pathways.

Radionuclide concentrations in arid soils are often strongly related to soil particle size with higher concentrations appearing in the smaller size fractions including silts and clays. This is an especially important distribution relationship because wind and water erosion processes

preferentially sort and transport the smaller soil particles compared to coarser fractions. This leads to an cnrichment of fine particles, often containing highest radionuclide concentrations, in eroding soil. The transport of fine soil particles, with higher radionuclide concentrations, followed by deposition on plant and animal surfaces, inhalation, and ingestion, serve as primary pathways for radionuclide entry into biota in these dusty, dry, environments.

The continued importance of physical transport of long-lived radionuclides in arid ecosystems will ultimately depend on the changes that occur in the distribution of the radionuclides in soil and sediment. At present, variable but significant amounts of the residual radionuclide inventory in most contaminated sites still occurs near the ground surface where erosional processes operate. Ultimately, the fate of long-lived radionuclides tightly bound to soil in arid environments, will depend on the fate of the soils and sediments themselves.

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